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Cincinnati, OH

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SEATTLE DISTRIBUTION SYSTEM CORROSION CONTROL STUDY

Volume V. Counteractive Effects of Disinfection and Corrosion Control

by

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Grant No. R 806686 010

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16. ABSTRACT

This study consisted of three research phases designed to evaluate the counteractive effects of corrosion treatment (pH adjustment) and disinfection (chlorination): (1) Electrochemical Tests - Copper corrosion rates were measured under varying pH, free chlorine residual and chloride concentration using a rotating disc electrode and linear potential sweep technique with solution resistance compensation. (2) On-Site Treatment Tests - The effects of simulated corrosion treatment start-up on chemical and microbial water quality from an old galvanized plumbing system were monitored. (3) Bacteriological Tests - The survival of Pseudomonas aeruginosa, Escherichia coli and Enterobacter aerogenes was determined under varying pH and free chlorine residual.

The results of the electrochemical tests showed an increase of approximately 70% in copper corrosion rates when free chlorine residuals were increased from 0 to 1.0 mg/L. These results indicate that increases in chlorine dosages may offset the corrosion reduction benefits realized with pH adjustment.

In the on-site treatment tests, standing water samples collected after treatment start-up displayed increased bacterial populations compared to untreated standing water samples. Iron-oxidizing and sulfate-reducing organisms were found in both standing water and tubercular incrustations. Opportunistic pathogens and coliform antagonists, including Pseudomonas and Flavobacterium, were detected in standing water samples, increasing in number with treatment start-up.

The results of the bacteriological tests showed a slower death rate at pH 8 for all three bacteria than at pH 7 or pH 6. However, at all the pH values tested, 100% mortality was achieved for all three species with a free chlorine residual of 0.2 mg/L or less and 1 minute contact time. The Seattle Water Department's present chlorination practices therefore should be sufficient to provide adequate disinfection at increased pH's.

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FOREWORD

The U.S. Environmental Protection Agency was created because of increasing public and government concern about the dangers of pollution to the health and welfare of the American people. Noxious air, foul water, and spoiled land are tragic testimonies to the deterioration of our natural environment. The complexity of that environment and the interplay of its components require a concentrated and integrated attack on the problem.

Research and development is that necessary first step in problem solution; it involves defining the problem, measuring its impact, and searching for solutions. The Municipal Environmental Research Laboratory develops new and improved technology and systems to prevent, treat, and manage wastewater and solid and hazardous waste pollutant discharges from municipal and community sources, to preserve and treat public drinking water supplies, and to minimize the adverse economic, social, health, and aesthetic effects of pollution. This publication is one of the products of that research and provides a most vital communications link between the researcher and the user community.

This report deals with the counteractive effects of disinfection (chlorination) and corrosion control (pH/alkalinity adjustment) in an aggressive water supply. The data indicate that although substantial corrosion reduction can be achieved by treatment, chlorination increases the aggressiveness of water, and corrosion treatment decreases the effectiveness of disinfection.

Francis T. Mayo
Director
Municipal Environmental Research
Laboratory

ABSTRACT

A study was conducted in Seattle, Washington, to evaluate the counteractive effects of corrosion treatment (pH adjustment) and disinfection (chlorination). The study consisted of three phases: (1) Electrochemical tests, which measured the copper corrosion rates under varying pH, free chlorine residuals and chloride concentrations using a rotating disc electrode and linear potential sweep technique with solution resistance compensation; (2) bacteriological tests, which determined the survival of *Pseudomonas aeruginosa*, *Escherichia coli*, and *Enterobacter aerogenes* under varying pH and free chlorine residuals; and (3) onsite treatment tests, which monitored the effects of simulated corrosion treatment start-up on chemical and microbial water quality in an old galvanized plumbing system.

The results of the electrochemical tests showed a substantial increase in copper corrosion rates with increased free chlorine residuals (approximately a 70% increase from 0 to 1.0 mg/L free chlorine). These results indicate that increases in chlorine dosages may offset the corrosion reduction benefits realized with pH adjustment.

The results of the bacteriological tests showed a slower death rate at pH 8 for all three bacteria than at pH 7 or 6. But 100% mortality was achieved at all the pH values tested for all three species with a free chlorine residual of 0.2 mg/L or less and a 1-minute contact time. The Seattle Water Department's (SWD) present chlorination practices should therefore be sufficient to provide adequate disinfection at increased pH, and SWD should not increase chlorine dosage upon implementation of the corrosion treatment program.

In the onsite treatment tests, standing water samples collected after treatment start-up displayed increased iron deposits, organic debris, and bacterial populations compared with untreated standing water samples. Iron-oxidizing and sulfate-reducing organisms were shown to exist within the pipe loop system in both the standing water and tubercular incrustations. Opportunistic pathogens and coliform antagonists, including *Pseudomonas* and *Flavobacterium*, were detected in standing water samples and increased in number with treatment start-up. The public health significance of sessile microbial communities in drinking water is also discussed.

This report is the fifth in a six-volume study of corrosion control in the Seattle, Washington, water distribution system. It was submitted in fulfillment of Grant No. R 806686 010 by the Seattle Water Department under the sponsorship of the U.S. Environmental Protection Agency. This report covers the period from January 1980 to December 1981, and work was completed as of May 1982.

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ABBREVIATIONS AND SYMBOLS

ABBREVIATIONS

A	-- surface area
A.A.S.	-- atomic absorption spectrophotometry
b	-- tafel slope
^oC	-- degrees Celcius
cm	-- centimeters
cm^2	-- centimeters squared
cor	-- corrosion reaction
C_β	-- bulk concentration of diffusing species
C_s	-- surface concentration of diffusing species
E^o	-- equilibrium potential
EDAX	-- energy dispersive X-ray analysis
EIS	-- environmental impact statement
EPA	-- U.S. Environmental Protection Agency
F	-- Faraday constant
$[\text{Halogen} + SO_4^{=}]/\text{Alk}$	-- $[\text{halogen + sulfate}]$/ alkalinity ratio
i	-- current density
I	-- current magnitude
J	-- mass transport rate
K	-- kinetic rate constant
L	-- liters
L/s	-- liters per second
MCL	-- maximum contaminant level
Me	-- metal
mg/L	-- milligrams per liter
m^3/d	-- cubic meters per day
n	-- electron equivalents
ox	-- oxidation reaction
re	-- reduction reaction
Re	-- Reynolds number
RPM	-- revolutions per minute
SEM	-- scanning electron microscopy
SPC	-- standard plate count
SWD	-- Seattle Water Department
V	-- reaction velocity
yr	-- year
	-- boundary layer
	-- kinematic viscosity
µg/L	-- micrograms per liter
µm	-- micrometers
µmhos	-- micromhos
	-- rotation speed

SYMBOLS

C	-- carbon
$CaCO_3$	-- calcium carbonate
CaO	-- calcium oxide (lime)
Cd	-- cadmium
CO_2	-- carbon dioxide
Cu	-- copper
e^-	-- electron
Fe	-- iron
H^+	-- hydrogen ion
HCO_3^-	-- bicarbonate
H_2O	-- water
Na_2CO_3	-- sodium carbonate
O_2	-- oxygen
pH	-- $-\log \{H^+\}$

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SECTION 1

INTRODUCTION

This report is the fifth in a series of six volumes describing the results of research conducted under the Seattle Distribution System Corrosion Control Study. Presented here are the results of three studies looking at the counteractive effects of disinfection (chlorination) and corrosion control (pH/alkalinity adjustment) in the Tolt distribution system. Volumes I and II present pilot test results and recommended specific chemical treatment for internal corrosion control in the Cedar and Tolt distribution systems, respectively. Volume III evaluates the potential of drinking water contamination from the corrosion of tin/antimony solder. Volume IV details the results of an onsite evaluation of corrosion treatment. And Volume VI presents a laboratory electrochemical technique for corrosion rate determinations in drinking water.

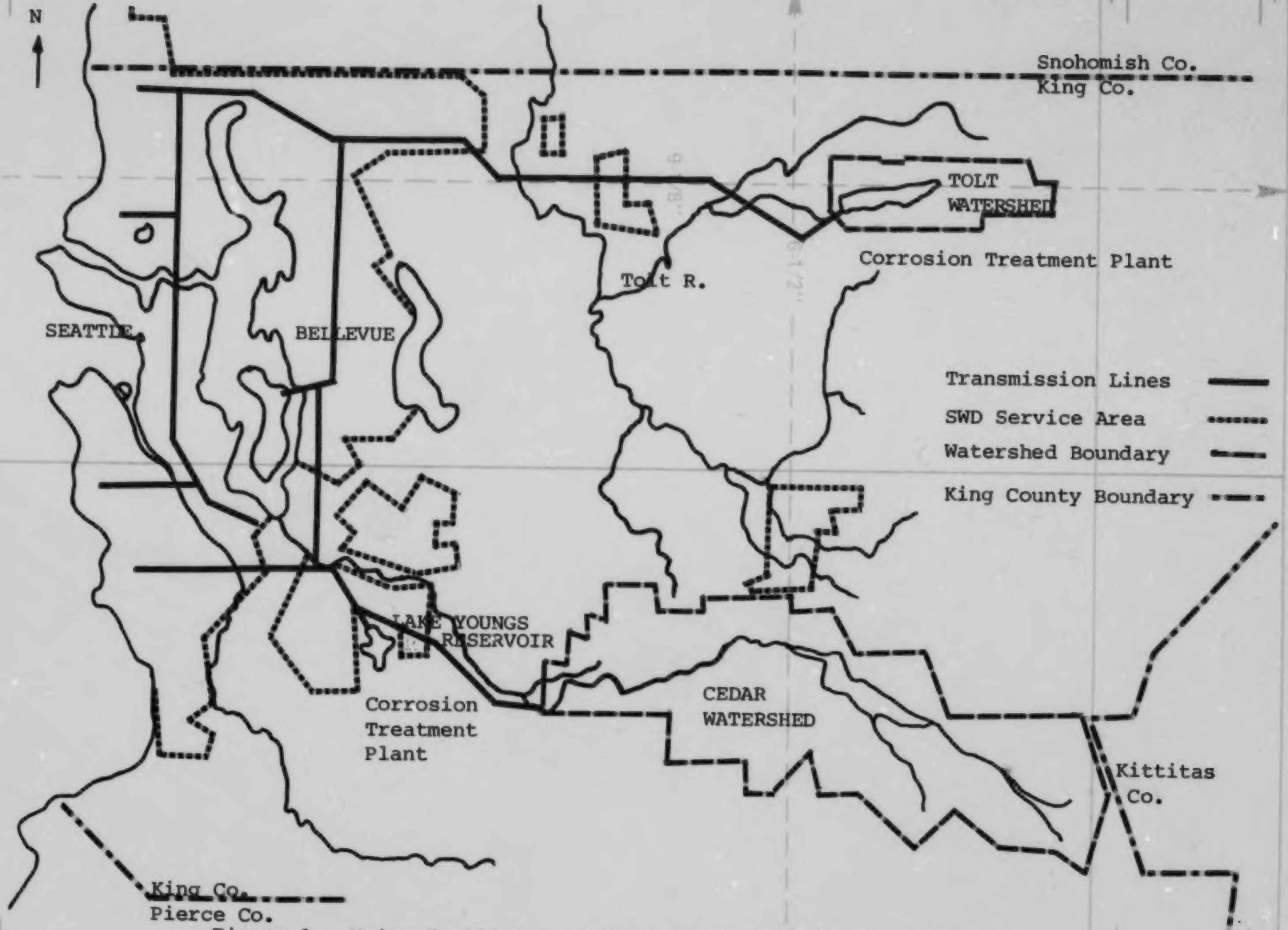
BACKGROUND

Water Sources

The City of Seattle is supplied with water from two sources - the Cedar and the Tolt Rivers, which furnish mountain water of unusual clarity and purity. The Cedar River system has been in operation since 1901 and the Tolt River system since 1964. Nearly 1 million persons are served directly or indirectly by the Seattle Water Department (SWD). Figure 1 illustrates the major facilities of the Seattle water distribution system.

Causes of Corrosion

Although the quality of both the Cedar and Tolt water supplies is high, both are corrosive to premise plumbing systems. Currently, the treatment of both water supplies consists of intake screening, chlorination, and fluoridation. The corrosiveness of the water results from several related factors, including (1) acidity, as indicated by low pH (after chlorination and fluoridation, pH is 6.8 to 7.2 for the Cedar supply and 5.8 to 6.2 for the Tolt supply); (2) dissolved oxygen concentration that is at saturated conditions; (3) insufficient calcium and bicarbonate alkalinity in the water to form protective calcium carbonate films on pipe surfaces; and (4) a relatively high [halogen + sulfate] /alkalinity ratio [(Halogen + $SO4^{=}$)/Alk] (0.5 to 0.8 for the Cedar supply and 2.5 for the Tolt supply), which is an indicator of conditions favorable to pitting corrosion.



N
Snohomish Co.
King Co.
TOLT WATERSHED
Corrosion Treatment Plant
Tolt R.
SEATTLE
BELLEVUE
Transmission Lines
SWD Service Area
Watershed Boundary
King County Boundary
LAKE YOUNGS RESERVOIR
Corrosion Treatment Plant
CEDAR WATERSHED
Kittitas Co.
King Co.
Pierce Co.

Figure 1. Major facilities of the Seattle distribution system.

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In 1970, three factors combined to intensify the corrosiveness of both water supplies:

1. The chlorine dosage at the open reservoir outlets was increased to decrease the occurrence of positive bacteriological samples within the distribution system.

2. Ammoniation of the water supply was stopped at the request of the U.S. Public Health Service to enable a free chlorine residual to be maintained throughout the distribution system. This change from combined to free chlorination was implemented to provide quicker, more effective disinfection of the unfiltered water supply.

3. Fluoridation with hydrofluosilicic acid began in 1970 based on a vote of the Seattle citizens in 1968.

Previous Studies Addressing the Corrosion Problem

Water quality deterioration in premise plumbing systems in a major concern of the SWD. Several projects that have examined the corrosivity of its water supplies have well documented that corrosion, although not a major concern in the distribution system, causes several problems in premise plumbing. In addition to causing the obvious aesthetic problems of rusty water, red and blue staining, and metallic tastes, plumbing corrosion is responsible for a sizable economic burden to Seattle homeowners and possibly some long-term health impacts to those with high environmental exposure to lead, cadmium, and copper.

In December 1975, the City of Seattle retained a consulting engineering firm to perform a detailed analysis of the corrosion problem and to recommend possible solutions. The Internal Corrosion Study,[1] which included a 9-month pilot-plant investigation, confirmed the corrosiveness of Seattle water, the causes of corrosion, and the impacts associated with the corrosive water. The study also evaluated alternative measures to reduce the corrosiveness of the water supply. Corrosion reduction alternatives investigated included alternative methods of disinfection and fluoridation, blending with groundwater supplies, and the addition of corrosion inhibitor chemicals.

Based on the findings of the consultant study and the conclusions of pilot plant investigations, an Internal Corrosion Control Management Plan was developed. Because the major causes of the water's corrosiveness were the very low levels of mineral solids, alkalinity, and saturation with dissolved oxygen, the proposed plan was designed to correct the natural deficiency of minerals in Seattle's water. The natural deficiency of calcium, silicate, and carbonate/bicarbonate alkalinity ions was proposed to be corrected by the addition of lime and sodium bicarbonate to both supplies, and possibly sodium silicate to the Tolt supply.

The Cedar and Tolt pilot plant studies were then conducted to define precisely the chemical treatment and dosages needed for corrosion control in both water supplies, and to document further the effects of such treatment.[2, 3]

These studies recommended the addition of lime only to the Cedar supply and lime plus sodium carbonate to the Tolt supply for internal corrosion control. The treatments were designed to achieve the water quality characteristics listed in Table 1. The recommendations were based on the overall corrosion protection each treatment affords to commonly used plumbing materials and the treatment cost and availability of chemicals.

SCOPE OF THE PROBLEM

Both the Internal Corrosion Study and the Cedar and Tolt Pilot Plant Studies recommended further investigation of the counteractive effects of chlorination and pH adjustment for corrosion control. The effect of pH adjustment on disinfection was clearly shown in a study by Butterfield.[4] In this study, the effectiveness of free chlorination was measured under various pH and temperature conditions for 11 strains of bacteria. The conclusions drawn from the study were that the killing efficiency of chlorine decreases with increasing pH and increases with increasing temperature and contact time.

Since the disinfection efficiency of free available chlorine residual decreases significantly as the pH rises, the SWD may have to increase chlorine dosages to provide adequate disinfection when pH adjustment is implemented. The addition of larger chlorine dosages may, however, offset the corrosion control achieved by pH adjustment. Several studies have shown that increases in chlorine residuals substantially increased internal corrosion.[5, 6, 7]

STUDY OBJECTIVES

The objectives of this research effort were to:

(1) Document the effects of free chlorine residual on corrosion rates.

(2) Determine whether chlorine dosages should be increased when pH adjustment is implemented to provide adequate disinfection.

TABLE 1. PRESENT AND PROPOSED WATER QUALITY CHARACTERISTICS

	Present Quality of Chlorinated and Fluoridated Water		Proposed Water Quality After Corrosion Treatment	
Parameter	Cedar	Tolt	Cedar	Tolt
pH	6.8-7.4	5.8-6.2	7.8-8.3	7.8-8.3
Alkalinity (as $CaCO_3$, mg/L)	15-18	2	20	14
(Halogen + Sulfate)/ Alkalinity Ratio	0.5-0.8	2.5-4.5	0.5	0.5

(3) Document the bacterial effects of the corrosion treatment program on water quality.

STUDY DESIGN

The study was divided into the following research phases:

Electrochemical Tests

Electrochemical corrosion rate determinations were conducted for copper in Cedar River water under various pH, free chlorine residual, and chloride concentrations to determine the effects of chlorination on corrosion rates. Corrosion rate determinations were made using a standard linear potential sweep method modified to include positive feedback compensation for potential drop resulting from solution resistance and the use of a rotating disc electrode for the elimination of mass transport limitations.

Microbiological Laboratory Tests

Microbiological laboratory tests were conducted to determine whether chlorine dosages should be increased upon implementation of corrosion treatment. The tests measured the effects of various pH and free chlorine residuals on the survival of *Pseudomonas aeruginosa*, *Escherichia coli*, and *Enterobacter aerogenes*. The tests were conducted at three free chlorine residuals and three pH values.

Onsite Corrosion Treatment Tests

During the period January 1981 to August 1981, tests designed to simulate corrosion treatment start-up were conducted at the City of Seattle Fire Station No. 35. This test site was chosen because it contained older galvanized plumbing that experiences severe corrosion-related water quality problems. Also, the test site was located in the Tolt distribution system where the changes in water quality caused by corrosion treatment will be greater than in the Cedar distribution system.

The corrosion tests documented the effects of corrosion treatment start-up based on metal leaching, bacteriological water quality, aesthetic water quality, and pipe tuberculation. Running water samples and 24-hour standing samples were collected from an 8.5-m pipe loop system. The samples were collected before treatment to establish baseline data and during the simulated treatment start-up.

SECTION 2

CONCLUSIONS

THE EFFECTS OF CHLORINATION ON CORROSION RATES

The electrochemical tests demonstrated that chlorination, using either NaOCl or Cl_2, substantially increases copper corrosion rates. A 1.0-mg/L free chlorine residual increased corrosion rates by approximately 20%. Also, for free chlorine concentrations greater than 0.2 mg/L, the corrosion rate at pH 8 was approximately equal to the corrosion rate at pH 7. Although previous studies showed that substantial corrosion reduction can be achieved by treatment to pH 8.0, the electrochemical test results indicate that increases in chlorine dosages would diminish the benefits received by pH adjustment.

THE IMPACT OF CORROSION TREATMENT ON DISINFECTION

The results of the microbiological laboratory tests demonstrate that the SWD will not be required to increase chlorine dosages to provide adequate disinfection when corrosion treatment is implemented. At pH 8 and 0.2 mg/L free chlorine residual, 100% kill for P. aeruginosa and E. coli was achieved with contact times of less than 30 seconds, and for E. aerogenes, with contact times of less than 2 minutes. The SWD chlorination program, which maintains a free chlorine residual of 0.6 mg/L for a minimum contact time of 10 minutes, will therefore provide a safety factor sufficient to insure adequate disinfection at pH 8.0.

THE EFFECTS OF CORROSION TREATMENT ON WATER QUALITY AT THE CUSTOMER'S TAP

The changes that will occur in overnight standing and running water quality at the customer's tap with corrosion treatment will vary depending on the parameter under consideration. The 24-hour standing water standard plate counts exhibited an increase of two to three orders of magnitude in microorganisms received per milliliter of sample compared with those counts received from the running water samples. Standing water samples collected after initiation of treatment displayed increased iron deposits, organic debris, and bacterial populations compared with untreated standing water samples. Iron-oxidizing organisms (e.g., Gallionella) and sulfate-reducing organisms (e.g., Desulfovibrio) were shown to exist within the pipe loop system in both the standing water and tubercular incrustations. Both of these organisms increased with corrosion treatment. Stalk- or bud-producing microorganisms (e.g., Caulobacter spp. and other iron-precipitating bacteria) were observed in the water samples and attached to the surface of, or embedded in, the tubercles. Pseudomonas aeruginosa counts increased 57% in standing water samples during corrosion treatment. Opportunistic pathogens and coliform antagonists including Pseudomonas spp. and Flavobacterium spp. were detected in standing water samples, increasing in number with treatment start-up. Opportunistic pathogens constituted approximately 77% of the total SPC isolates in 24-hour standing water samples, whereas coliform antagonists accounted for approximately 71% of the total SPC isolates.

In running water, metal reductions occurred in zinc leaching and iron leaching during treatment start-up. No appreciable change occurred in lead leaching, cadmium leaching, apparent color, and turbidity concentrations in running water.

In standing water, zinc leaching was reduced during treatment from pH 6 to 7 and increased from pH 7 to 8. Iron leaching increased by approximately 35% during treatment start-up, and copper and lead leaching were decreased 53% and 57%, respectively. Also, apparent color and turbidity increased in standing water during treatment start-up.

SECTION 3

RECOMMENDATIONS

SEATTLE WATER DEPARTMENT

The SWD should:

- Implement the corrosion treatment start-up procedures mentioned in this report and detailed in Volume IV, On-Site Evaluation of Corrosion Treatment.
- Continue with present chlorination practices and maintain chlorine dosages when corrosion treatment is implemented.
- Monitor bacteriological water quality in the distribution system before and after the implementation of full-scale treatment.
- Evaluate the possibility of trihalomethane formation in Tolt and Cedar water with respect to chemical treatment.
- Determine whether corrosion treatment will cause post-precipitation problems in the distribution system.

U.S. ENVIRONMENTAL PROTECTION AGENCY

The U.S. Environmental Protection Agency (EPA) should:

- Publish a state-of-the-art manual for corrosion control of drinking water.
- Investigate more fully the impact of chlorination practices on the corrosion of household plumbing materials.

SECTION 4

EQUIPMENT DESIGN AND OPERATION

ELECTROCHEMICAL TESTS

Overview

Electrochemical methods have been used in corrosion research for nearly twenty-five years, but only in the last decade have these methods been developed on a quantitative basis for use in drinking water systems, especially for waters of low conductivity (< 150 µmhos/cm). The principal advantage of electrochemical methods is that a corrosion rate determination can be made rapidly and an instantaneous rate is measured, as opposed to a rate averaged over a period of several months as in a weight loss coupon test. Rapid and instantaneous corrosion rate measurements are useful in observing the change in corrosion rate that occurs with slight changes in solution composition, an important consideration in this research.

Instrumentation

The basic equipment required to generate a polarization curve consists of an electrochemical cell, a potentiostat to control electrode potential, a millivoltmeter to measure electrode potential, and a microammeter to measure cell current. The basic electrochemical cell is composed of three electrodes, the working electrode where the reaction rate measurement occurs, a reference electrode which along with a luggin capillary allows measurement of electrode potential of the working electrode, and a counter electrode which provides a reaction site for the half-cell reaction not being measured. The cell current which flows between the working and counter electrodes is measured with a microammeter. Electrode potential is measured with a millivoltmeter as the potential drop between the working and reference electrodes. A simple experimental system which can be used to generate polarization curves has been published in an ASTM Standard[8] and is shown in Figure 2. By manually changing the electrode potential of the working electrode with a potentiostat and recording the cell current that flows at each electrode potential value, one can generate a polarization curve. This set-up is similar but not the actual experimental apparatus used on this project. It is presented here for the purpose of illustrating the basic manner in which a polarization curve is generated.

The experimental apparatus used in this study contains the same essential electrochemical cell and potentiostat set-up shown in Figure 2. Additional instrumentation used include a function generator (to sweep electrode

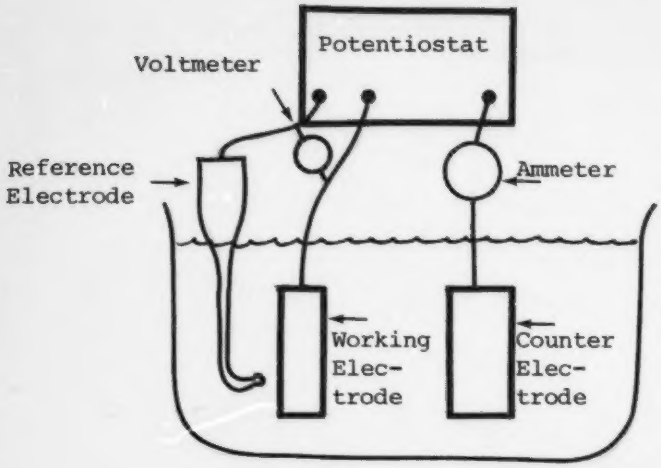

Potentiostat
Voltmeter
Reference Electrode
Ammeter
Working Electrode
Counter Electrode

Figure 2. ASTM standard electrochemical cell for corrosion tests.

potential linearly with with respect to time), an X-Y recorder (to record the plot of cell current versus electrode potential), an IR Compensator (which allows removal of solution resistance effects in low conductivity water), and a rotating disc electrode (which allows measurements to be made under conditions of either kinetic or transport rate control). A schematic diagram of the instrumentation used is shown in Figure 3.

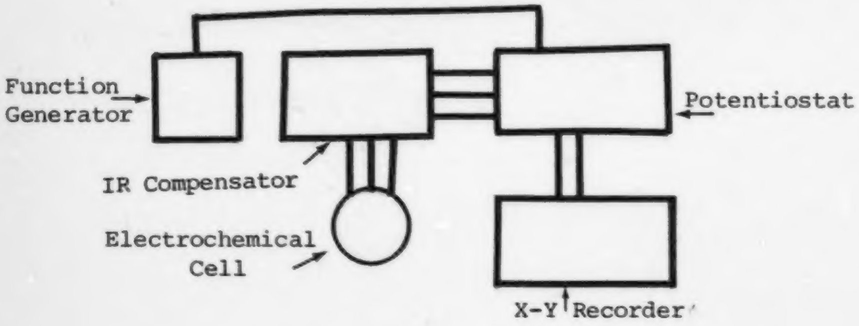

Function Generator
Potentiostat
IR Compensator
Electrochemical Cell
X-Y Recorder

Figure 3. Experimental apparatus.

This set-up allowed a somewhat automated polarization curve generation which tended to eliminate differences in operating procedures between runs and enhance reproducibility. A detailed description of each component of this apparatus can be found in Volume VI of the Seattle Distribution System Corrosion Control Study.[9]

ONSITE TREATMENT TESTS

Test Site and Equipment

The onsite treatment tests were conducted at the City of Seattle Fire Station No. 35, which is located in the Tolt distribution system in the north end of Seattle. The flow schematic of the onsite treatment plant is illustrated in Figure 4. As the water entered the fire station, it passed through the chemical feeding and mixing system (Figure 5) and into the existing building plumbing system. Attached to the end of the existing plumbing system was the 1.9-cm-diameter old galvanized pipe test loop. The pipe loop system was made from 10-year-old galvanized hot water pipe which was collected from the University of Washington. At the time of installation of the pipe loop, the pipe was highly tuberculated. The pipe loop was 8.5 m in length and contained 5 elbows. Pressure test gauges were located at the beginning and end of the loop for determination of headloss in the loop. A flow of 10.5 L/min through the loop was maintained using a throttling valve located at the end of the loop was was measured by a flow meter also at the end of the loop. The throttling valve was periodically shut off for the collection of 24-hour standing samples.

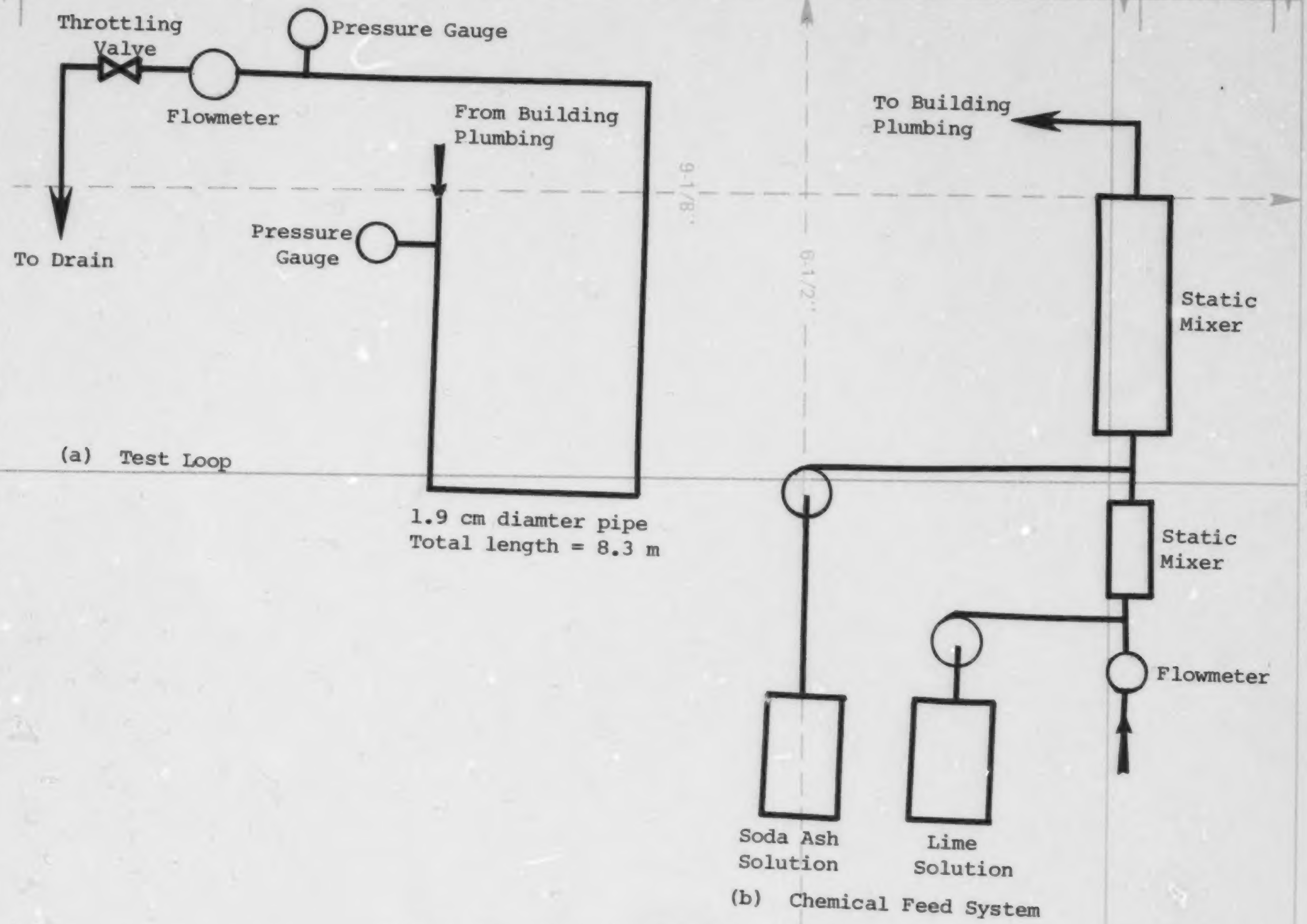
The chemical feeding and mixing system is illustrated in Figure 5. The chemicals were fed by two Liquid Metronix, Inc., diaphram pumps. The chemicals used were hydrated lime [$Ca(OH)_2$] and sodium carbonate [$NaCO_3$]. Feed solutions of the full strength chemicals [170 g $CaOH_2$ / 151 L; 860 g Na_2CO_3 / 151 L] were mixed in 151 L plastic buckets. The feed solutions were then pumped from the solution buckets into the plumbing system just before the static mixers.

The chemical feed pumps were paced proportionally to the main flow volume using a Signet pump pulser. Flow was measured using a Signet Paddle Wheel Flow Sensor and self-powered flow meter.

Chemical Treatment

The test treatment conditions were chosen to simulate average distribution system levels of pH and alkalinity during full-scale treatment start-up. Table 2 presents the target water quality for the test conditions.

The treatment phases listed in Table 2 were implemented in succession to simulate treatment start-up. Phase I, which lasted 106 days, was used to collect baseline data. Phase II (37 days), Phase III (18 days) and Phase IV (27 days) simulated the gradual increases in pH and alkalinity which will occur during full-scale treatment start-up.


Throttling Valve
Pressure Gauge
Flowmeter
From Building Plumbing
Pressure Gauge
To Drain
1.9 cm diamter pipe
Total length = 8.3 m
To Building Plumbing
Static Mixer
Static Mixer
Flowmeter
Soda Ash Solution
Lime Solution

(a) Test Loop

(b) Chemical Feed System

Figure 4. Pilot plant flow schematic.

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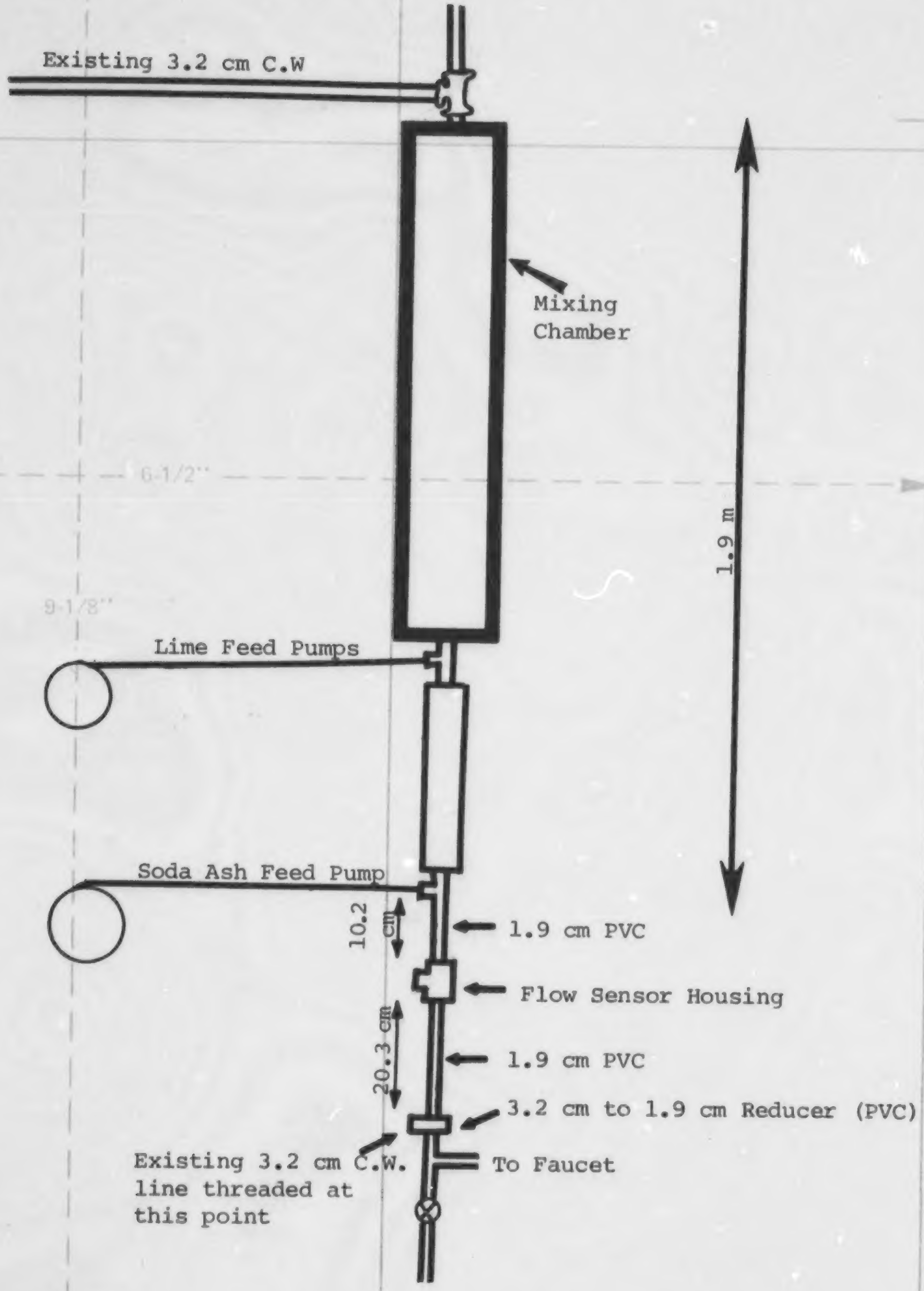
Existing 3.2 cm C.W
Mixing Chamber
1.9 m
Lime Feed Pumps
Soda Ash Feed Pump
10.2 cm
1.9 cm PVC
Flow Sensor Housing
20.3 cm
1.9 cm PVC
3.2 cm to 1.9 cm Reducer (PVC)
Existing 3.2 cm C.W. line threaded at this point
To Faucet

Figure 5. Pilot plant mixing system.

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TABLE 2. ONSITE TEST TREATMENT CONDITIONS

Item	Phase I	Phase II	Phase III	Phase IV
pH	6.00	7.00	7.60	8.00
Alkalinity (mg/L $CaCO_3$)	2.0	9.0	8.0	14.0
Lime Dosage, CaO (mg/L)	0	0	2.00	1.70
Na_2CO_3 Dosage (mg/L)	0	7.42	6.40	9.00

Scanning Electron Microscopy

Scanning electron microscopy was heavily employed (yielding 309 micrographs) for identification of microorganisms in the water samples and examination of tubercular incrustations from the interior pipe loop surface. X-ray energy-dispersive microanalysis was used to determine the elemental composition of particulate matter in water samples as well as the chemical make-up of the tubercle.

Scanning electron microscopy was performed by the Materials Analysis Center of the University of Washington's Department of Mining, Metallurgical and Ceramic Engineering.

The Center has a Cambridge Mark IIA Scanning Electron Microscope (SEM) capable of 200 Angstrom resolution, which was used in the secondary electron emission mode. The instrument has useful magnification to 50,000X and is unique in that its large depth of focus allows unusual observation and photography of three-dimensional structures not possible with other microscopic techniques.

The SEM is equipped with the EDAX Energy Dispersive X-Ray Detector for elemental analysis from fluorine to uranium in the atomic table. Qualitative analysis in the SEM mode was conducted with spectra generation typically less than a minute.

SECTION 5

ANALYSIS AND PROCEDURES

ELECTROCHEMICAL TESTS

Fundamentals of Electrochemical Polarization

Electrochemical methods can be used to measure corrosion rates in aqueous systems when oxidation-reduction reactions are involved. Consider the simple pair of redox half-cell reactions shown below:

$$Me \xrightarrow{kox(E)} Me^{+1} + e^{-}$$

$$1/4\ O_2 + H^{+} + e^{-} \xrightarrow{kre(E)} 1/2\ H_2O$$

Since each half-cell reaction involves the transfer of electrons, the rate at which electrons are transferred is a measure of the overall reaction rate, the corrosion rate. This rate can be measured experimentally as a current density ($\mu A/cm^2$) by use of an electrochemical cell and the corrosion rate can be calculated using the Stearn-Geary[10] equation:

$$i = i_{cor} \left[\exp\left(\frac{E - E_{cor}}{ba}\right) - \exp\left(- \frac{E - E_{cor}}{bc}\right) \right]$$

This equation relates the net current density, i, flowing in an electrochemical cell with the electrode potential, E, of the working electrode. Both of these parameters, i and E, are measurable, and if an experimental plot of i vs. E can be obtained, the values of i_{cor}, E_{cor}, b_a, and b_c can be evaluated. This is how corrosion rates are determined electrochemically. Although derived solely for reactions under kinetic rate control, the Stearn-Geary model has practical use in systems where one half-cell reaction is diffusion controlled, as well. The constants b_a and b_c, called anodic and cathodic tafel slopes, have numerical values characteristic of the particular half-cell reaction they represent.

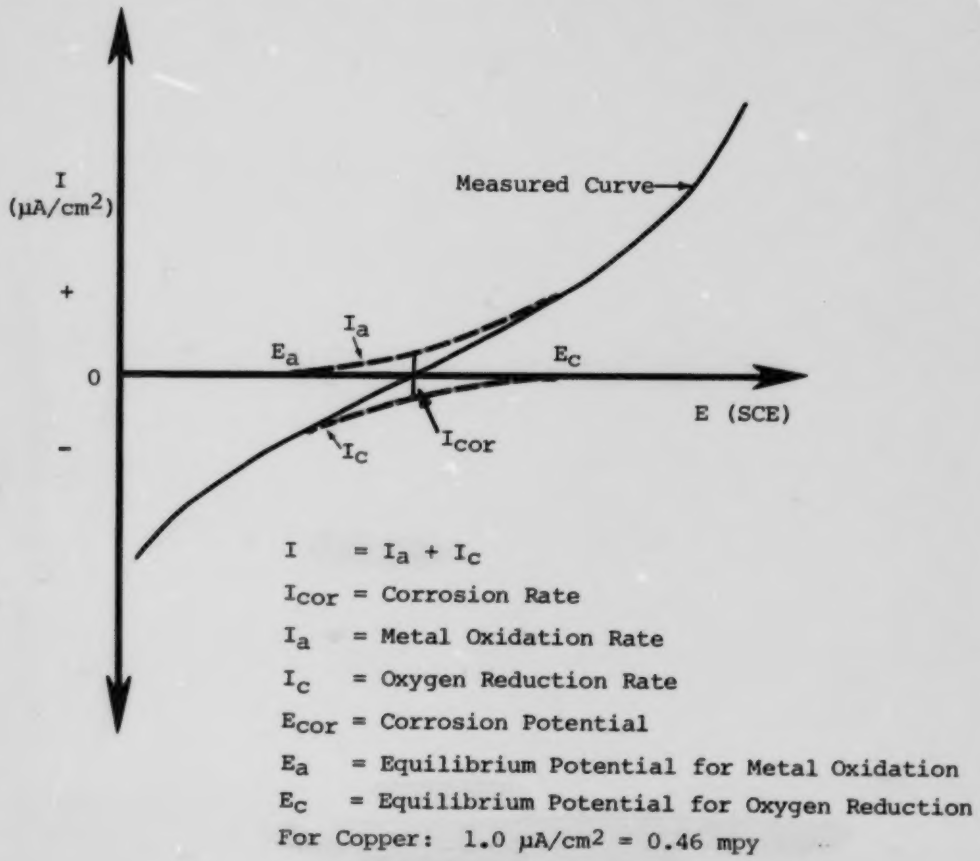
When the anodic half-cell is diffusion controlled, the value of $b_a \rightarrow \infty$ and the exponential term containing it approaches 1 in value. When the cathodic half-cell reaction is diffusion controlled, the value of $b_c \rightarrow -\infty$

and the exponential term containing it approaches 1 in value. In either case, the value of i_{cor} and the other tafel slope can still be determined from an i vs. E plot.

The experimental output of an electrochemical corrosion rate determination is a plot of net cell current density, i ($\mu A/cm^2$), vs. electrode potential, E (mv vs. SCE). The corrosion rate is then calculated from this plot called a polarization curve. Figure 6 shows a typical polarization curve representing a corroding system in which both half-cell reactions are proceeding under rate control by chemical kinetics.

Calculating Corrosion Rates from Polarization Curves

Corrosion rates were calculated from polarization curves using the 3-point method. This method, originally developed by Barnartt[11] for investigation of electrode reaction mechanisms, is directly applicable to corroding systems. The method is versatile in that it is applicable to electrode reactions where both half-cell reactions are proceeding under rate control kinetics or where one half-cell reaction is transport limited.



$I = I_a + I_c$

I_{cor} = Corrosion Rate

I_a = Metal Oxidation Rate

I_c = Oxygen Reduction Rate

E_{cor} = Corrosion Potential

E_a = Equilibrium Potential for Metal Oxidation

E_c = Equilibrium Potential for Oxygen Reduction

For Copper: 1.0 $\mu A/cm^2$ = 0.46 mpy

Figure 6. Steady-state polarization curve.

For the 3-point method, three data points are needed, two of them selected at potentials positive with respect to E_{cor} and one negative with respect to E_{cor}. The first data point is arbitrarily selected to be (E_1, I_1) where $(E_1 - E_{cor}) = \Delta E$. Data point number two (E_2, I_2) is then selected so that $(E_2 - E_{cor}) = 2\Delta E$ and data point number three (E_3, E_{cor}) so that $(E_3 - E_{cor}) = -2\Delta E$. Figure 7 shows graphically this method of data point selection.

Once the points are chosen, the current magnitudes, I, at the three points can be used to calculate the corrosion rate using the following relationship derived by Barnarrt.[12]

$$i_{cor} = \frac{Icor}{A} = \frac{I_1}{\left[\left| I_2/I_1 \right|^2 - 4 \left| I_2/I_3 \right|^{\frac{1}{2}} \right]^{\frac{1}{2}} A} \ (\mu A/cm^2)$$

where A is the surface area of the working electrode in cm^2. This method has been used to calculate corrosion rates for polarization curves taken under kinetic rate control. Corrosion rates calculated in ($\mu A/cm^2$) can be readily converted to mils per year.

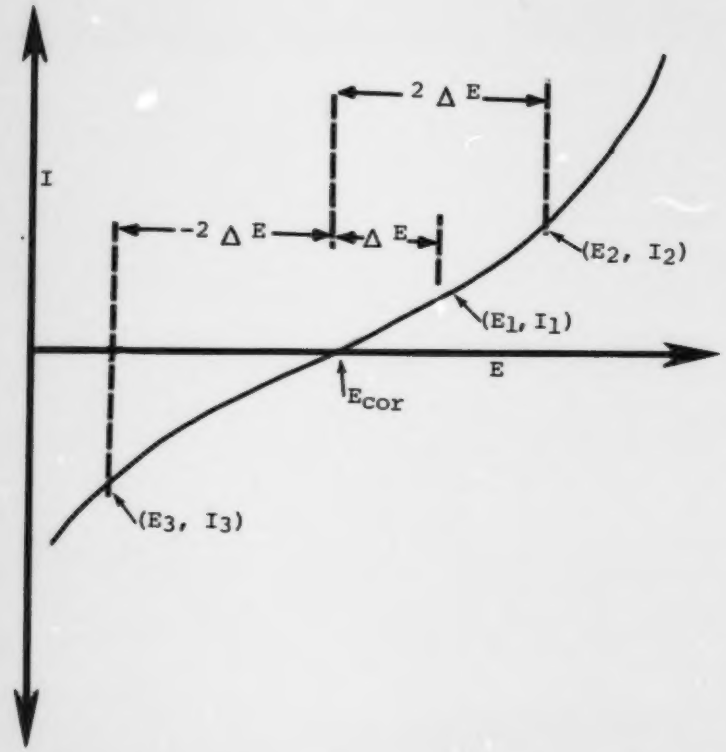


Figure 7. Graphical representation of the data point selection method.

Test Solution

Cedar River water, collected from Seattle's Beacon Hill Reservoir, was used for the electrochemical test. The water was collected in plastic cubetainers and was filtered through a Millipore filter upon arrival to the laboratory.

Table 3 lists the individual parameters varied in the electrochemical tests, the chemical used to vary the parameter and the range of values tested. Test solutions were made up immediately prior to corrosion rate measurement.

Corrosion Rate Measurement

When preparation of the electrodes, cell, and test solution was complete, along with pH and conductivity measurements on the test solution, the electrode cell was assembled by placing the electrodes in the test solution and hooking up the shielded electron leads that run between the electrodes and the potentiostat. A reducing (∽200 mv below E_{cor}) was placed on the working electrode to prevent any oxidation from occurring. The X-Y recorder was zeroed and calibrated and the appropriate IR compensation was dialed in for the upcoming test run. The potential scanner, set at the appropriate scan rate, was then turned on and the voltage scan began. The electrode potential of the working electrode was then followed, as it changed with respect to time, on the X-Y recorder which keeps a plot of electrode potential and the corresponding cell current. The potential scanner then sweeps the electrode potential of the working electrode in a positive direction through E_{cor} and on to a value ~150 mv positive of E_{cor}. The magnitude of the cell current starts off negative indicating that a net reduction reaction was occurring at the working electrode. Gradually this current was reduced to zero as the potential was swept through the corrosion potential. It then increases in the positive direction as the oxidation half cell takes over at potential values more positive than E_{cor}.

From the plot generated from this procedure, the corrosion rate was calculated which correspond to the composition of the test solution used in that run. By varying the concentration of a desired solution parameter

TABLE 3. SAMPLE WATER PREPARATION

Parameter Varied	Treatment Chemical	Range Tested
pH	$Ca(OH)_2$ and $CO_2(g)$	6.0 to 8.3
Free Chlorine Residual	Solution water from gas chlorinator	0-1.8 mg/L free chlorine
Free Chlorine Residual	NaOCl	0-1.8 mg/L free chlorine
Chloride	NaCl	3.4 to 11.0 mg/L

and making corrosion rate determinations at each concentration, the effects of that parameter on the corrosion rate was determined quantitatively.

Water Quality Measurements

pH and conductivity measurements were made on the test solution prior to and following each test run. On occasion pH was followed continuously throughout a run. pH was measured with a Metrohm/Brinkman Model: pH-104 Digital pH meter using a Beckman, Altex, ceramic junction, silver/silver chloride reference electrode. The pH meter was normally calibrated at least once daily at the temperature at which experimental runs were to be made. Calibration at pH 6.86 was found to be adequate since most measurements were made between $6.0 \leq pH \leq 9.0$. The test solution was stirred during pH measurement and a response time of roughly 10 minutes was found necessary for stabilization of pH in low conductivity water.

Conductivity was measured with a Barnsted Conductivity Bridge Model PM-70B, using a Barnsted Model E3421 conductivity cell with a 0.1 cell constant. The cell was directly immersible in the test solution and completely self-contained. The instrument required little calibration or maintenance, just a rinsing of the cell electrodes with distilled deionized water which was changed daily. The instrument response time was fairly rapid except at low conductivities (below ~30 μmhos/cm) when 8-10 minutes was needed for stabilization.

Chloride ion was determined by titration with mercuric nitrate according to the Mercuric Nitrate Method in Standard Methods.[13]

Free chlorine residuals were determined after 10 minutes with a Fischer and Porter 17T2000 amperometric titrator, according to procedures given in the Fischer and Porter manual (21) and in Standard Methods.[13]

BACTERIOLOGICAL TESTS

Experimental Design

The bacteriological tests were conducted to determine whether SWD's present chlorination practices will provide adequate disinfection at the pH values achieved with corrosion treatment.

The experimental variables and the bacteria used are presented in Table 4. A separate test was performed for each combination of bacteria, water source, chlorine residual, pH, and exposure time.

Solution Water

Water was collected from Seattle's Lake Forest Park Reservoir (Tolt River water) and Beacon Hill Reservoir (Cedar River water). In the laboratory, chlorine concentration was determined iodometrically to insure that no chlorine residual was present in the reservoir water. The reservoir

TABLE 4. EXPERIMENTAL VARIABLES

Strain Bacteria	Water Source	Chlorine Residual (mg/L)	pH	Time of Exposure to Chlorine (minutes)
Pseudomonas aeruginosa	Tolt	0.1	6	0
				0.5
Escherichia coli	Cedar	0.2	7	1.0
Enterobacter aerogenes		0.1 (dosage)	8	2.0

water was also measured by the standard plate count method to get a background bacterial count.

The test solutions were made up by measuring 360 mL of reservoir water and 40 mL of the appropriate bacterial suspension into one L sterile glass bottles in order to give the test solution an initial background count of 20,000-70,000 organisms/mL. Tolt water was used for the tests conducted at pH 6 and Cedar water for the tests at pH 7. For the pH 8 tests, Tolt water was adjusted to pH 8 using a 2 N sodium carbonate solution.

Test Sequence

After the test solutions were prepared and thoroughly mixed, the initial pH and alkalinity was determined. After mixing, one mL aliquots of the test solution were aseptically removed and plated in triplicate to be used as the control sample for each bottle. A quantity of chlorine stock solution required to create the desired chlorine residual was then added to the remaining volume in each bottle. After mixing, one mL aliquots of the test solution were plated in triplicate at intervals of 30 seconds, 1 minute, and 2 minutes. All one mL aliquots were pipetted into petri dishes and immediately solidified with tryptone glucose yeast agar. Free chlorine residual and pH were then determined on each bottle of test solution after 10 minutes.

All agar plates were incubated at 35° ± 0.5°C for 48 ± 3 hours. The plates were then counted and the percentage of bacteria killed at each time interval was determined.

Analytical Techniques

Reagents --

All reagents were prepared according to *Standard Methods*[13] unless otherwise stated.

The stock chlorine solution was made to simulate a gaseous chlorine solution: 4.5% NaOCl bleach solution was diluted 1:100 and adjusted to pH

3.5 with 1 N HCl. This gave an approximate 500 mg/L Cl_2 solution.

Phenylarsine oxide (PAO), 0.01 N, was made by dilution of reagent grade 0.025 N PAO and standardized with 0.0282 N iodine.

A bicarbonate buffer, 0.5 N, was made from reagent grade sodium bicarbonate. Stock solutions of pH 7 and pH 10 buffer were used. The following reagents were made by dilution of reagent grade stock: sulfuric acid, 0.0200 N and sodium carbonate, 2 N.

Chemical Techniques --

The concentration of the stock chlorine solution was analyzed each day. Standardization was performed using the iodometric method; standard 0.100 N $KNIO_3$ was used to standardize the approximate 0.025 N $Na_2S_2O_3$.

Free available chlorine concentrations were determined with a Wallace and Tiernan WIA790-1-2 amperometric titrator, according to the procedures given in the Wallace and Tiernan manual[15] and in Standard Methods.[13] Negative response to chloramines was documented by preparing a monochloramine solution according to the 1969 Analytical Reference Service "Water Chlorine Residual No. 1 Report"[16]. In some cases, total chlorine residuals were performed by DPD colorimetry using either a HACH DPD kit or a B & L 710 Spectrophotometer. Alkalinity was determined initially at each pH level by the potentiometric method according to Standard Methods.[13]

Biological Techniques --

Pure cultures of *P. aeruginosa*, *E. coli*, and *E. aerogenes* were received from the University of Washington Department of Microbiology.

Portions of the cultures were washed and packed with 20 mL of sterile chlorine demand free water and sterile glass beads, decanted, and centrifuged at 12,000 rpm for 20 minutes.

The supernatant was discarded and the cells were rewashed, packed, decanted, and centrifuged two additional times. The washed cells were then resuspended in 100 mL of chlorine demand free water and the suspension was transferred to a 250 mL sterile glass bottle. The washed cells were stored in the refrigerator at 4°C.

After one week a standard plate count was made to determine the bacterial concentration of the suspension. A logarithmic dilution was made with sterile phosphate buffer, pH 7. One mL aliquots of the bacterial suspension were then removed and pipetted into a petri dish, and solidified with tryptone glucose yeast agar. Plates were incubated at 35 ± 0.5°C for 48 hours. From these counts, the bacterial concentration of the original suspension was determined.

ONSITE TREATMENT TESTS

Sample Collection

The effects of treatment start-up were determined from standing and running samples and headloss determinations collected from the test pipe loop system. Standing water samples represented 24-hour stagnant water conditions in the pipe loop system. Running water samples were collected after a two-mine flushing at 10.5 L/min of the pipe loop system following the collection of the 24-hour standing sample. Control samples were collected at the site where the distribution system water first entered the pilot plant.

Physical and Chemical Parameters

One liter (L) samples of 24-hour standing, running, and control water were collected after each stagnation period. From these samples, 250 mL aliquots were immediately preserved (0.5% HNO_3 in Pyrex) for metals analysis.

Water from the one liter samples was analyzed for alkalinity, conductivity, turbidity, apparent color, total suspended solids, and calcium concentration according to Standard Methods. The metals analyzed and analysis techniques for the 250 mL aliquots are presented in Table 5.

Microbiological Examination

Collection of Samples --

Water samples as well as tubercular incrustations were collected from the test pipe loop prior to corrosion treatment to determine the present microbial quality of Tolt distribution water in "old" galvanized premise plumbing. Regular monitoring of both these parameters was continued throughout treatment phases II, III and IV to observe any qualitative and quantitative changes occurring within the microbial population in the pipe loop due to corrosion treatment.

Water samples were collected twice per week in 1 liter sterile polyethylene bottles treated with 10% sodium thiosulfate to neutralize any chlorine residual in the water samples. The temperature of all water samples was determined upon collection with a SYBRON/Taylor 21438-1 pocket thermometer.

TABLE 5. METALS ANALYZED AND ANALYSIS TECHNIQUES

Metal Tested	Analysis Method
Fe, acid hydrolizable	Flame Atomic Absorption Spectrophotometry
Zn, " "	" " " "
Cu, " "	" " " "
Pb, " "	Flameless Atomic Absorption Spectrophotometry
Cd, " "	" " " "
Na, " "	Flame Emission Spectrophotometry

In general, most water samples were analyzed within five hours after collection. Occasionally, the samples were refrigerated at 4°C if analysis could not be completed immediately, but all were processed within 12-18 hours. Due to high levels of turbidity, most notably in the standing water samples, 3-5 drops of a sterile 1% Tween 80 solution (a surfactant) were added to each 1 liter sample bottle to minimize any bacterial clumping. The bottles were then thoroughly shaken 25 times in a 45°C arc for 15 seconds before dilutions were made.[17]

Microbial Differentiation --

Coliforms, standard plate count bacteria, and various other microorganisms of interest, for example, iron and sulfur transforming organisms, specifically iron-precipitating and sulfate-reducing bacteria, were cultured on various selective enrichment media. In addition to water samples, tubercles were also examined for the presence of the above-mentioned organisms. For inoculation, the tubercles were manually ground in a sterile mortar, the samples weighed, and the number of organisms recorded per gram of tubercle. A description of the various selective enrichment culture media used is presented in Appendix A.

Verification of microbial growth in various enrichment media was conducted by means of direct microscopic or scanning microscopic examination of cellular morphology, as well as Gram staining and biochemical testing.

Enumeration of coliforms--Total coliforms were enumerated by the membrane filtration (MF) technique (0.45 µm pore size membrane filter) and m-Endo LES agar.[17] Incubation was at 35 ± 0.5°C for 24 hours. Typical coliform colonies from the MF technique were verified by using lauryl tryptose broth (Difco Laboratories). Fecal coliforms were enumerated on m-FC medium (Difco) according to *Standard Methods*.[17]

Enumeration and identification of SPC bacteria--SPC bacteria were enumerated by the pour plate technique. Two different incubation times and temperatures were used: 1) Incubation at 35 ± 0.5°C for 48 ± 3 hours and 2) Incubation at 20-24°C for 6 days. Bacterial density of the water was calculated by enumerating the viable colonies on plates after each incubation with a Quebec colony counter.

A total of 2210 colonies were picked from the standard plate count agar. These isolates were purified by streaking onto agar composed of tryptic soy broth (Difco) supplemented with 0.3% yeast extract (TSB-YE; Difco) and 1.5% agar (Difco), then incubated at 35 ± 0.5°C for 48 ± 3 hours or 20-24°C for 6 days. The incubation time was determined based on the original incubation time of the isolate. Pure cultures were maintained on slants of TSB-YE agar at 4°C.

All isolates were placed into genera or groups by noting colony and cell morphology, catalase and oxidase reactions, motility, urease, indole production, anaerobic growth, and glucose fermentation or oxidation (Figures 8 and 9)[17]. The isolates were assigned to arbitrary numbered groups on the basis of the pattern obtained with these seven tests. Gram reaction, nitrate reduction, citrate utilization, and growth on MacConkey agar were determined

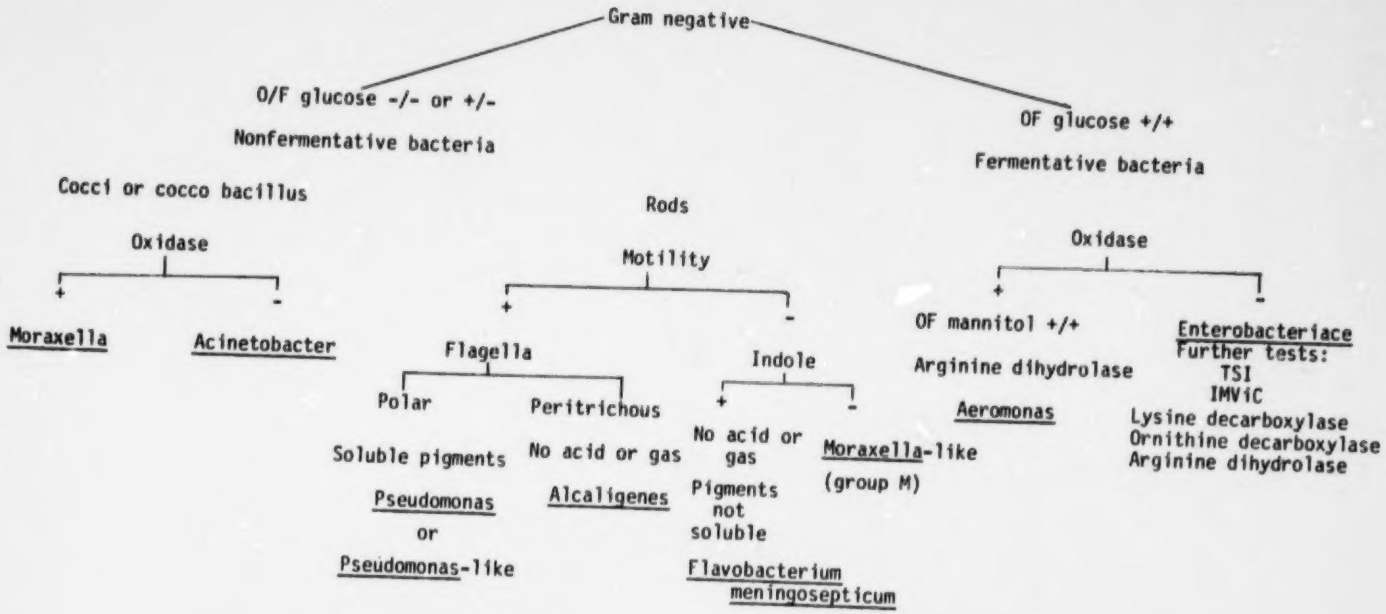

Gram negative
O/F glucose -/- or +/-
Nonfermentative bacteria
OF glucose +/+
Fermentative bacteria
Cocci or cocco bacillus
Oxidase
+
-
Moraxella
Acinetobacter
Rods
Motility
+
-
Flagella
Polar
Peritrichous
Soluble pigments
Pseudomonas
or
Pseudomonas-like
No acid or gas
Alcaligenes
Indole
+
-
No acid or gas
Pigments not soluble
Flavobacterium meningosepticum
Moraxella-like
(group M)
Oxidase
+
-
OF mannitol +/+
Arginine dihydrolase
Aeromonas
Enterobacteriace
Further tests:
TSI
IMViC
Lysine decarboxylase
Ornithine decarboxylase
Arginine dihydrolase

Figure 8. Proposed scheme for identification of Gram negative SPC bacteria from potable and raw water supplies.[17]

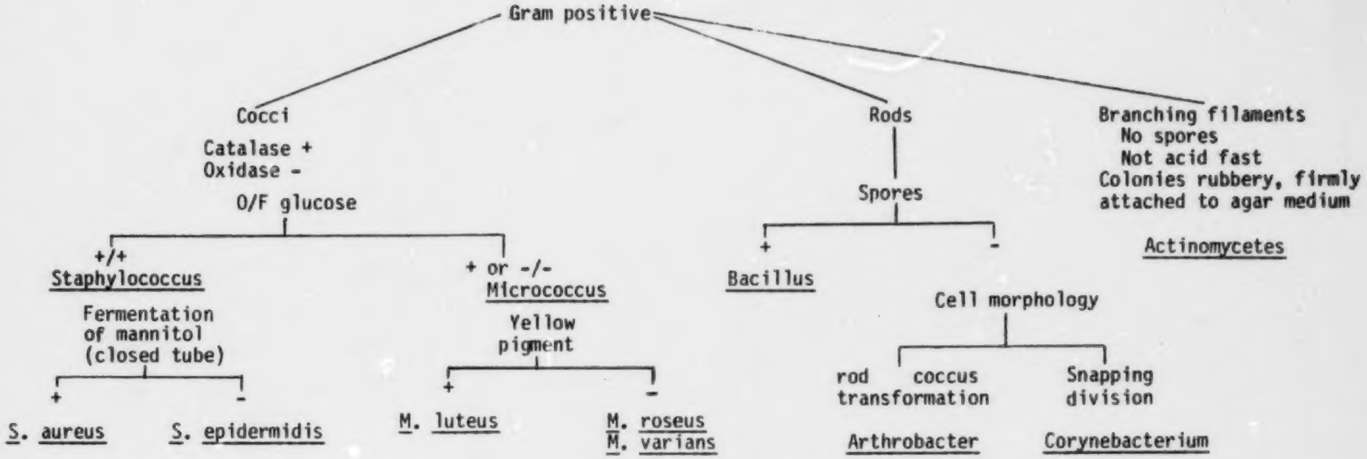

Gram positive
Cocci
Catalase +
Oxidase -
O/F glucose
+/+
Staphylococcus
Fermentation
of mannitol
(closed tube)
+
-
S. aureus
S. epidermidis
+ or -/-
Micrococcus
Yellow
pigment
+
-
M. luteus
M. roseus
M. varians
Rods
Spores
+
-
Bacillus
Cell morphology
rod coccus
transformation
Snapping
division
Arthrobacter
Corynebacterium
Branching filaments
No spores
Not acid fast
Colonies rubbery, firmly
attached to agar medium
Actinomycetes

Figure 9. Proposed scheme for identification of Gram positive SPC bacteria from potable and raw water supplies.[17] [a] excludes catalase negative streptococci.

for a portion of the isolates in each group to obtain further information and to evaluate homogeneity within the group. Media and reagents were prepared according to standard procedures (185). Hugh Leifson O/F medium with 1% glucose or 1% mannitol, methyl red Voges-Proskauer medium, Moeller decarboxylase broth, triple sugar iron (TSI) agar slants, Simmons citrate agar, MacConkey agar, nitrate agar, and urease test agar were prepared from Difco products. Lysine, ornithine, and arginine amino acids were obtained from Sigma Chemical Co. Triple sugar iron agar slants were incubated for 24 hours. The Voges-Proskauer test was performed after 48 hours of incubation.

Identification of Pseudomonas aeruginosa was performed using the membrane filtration (MF) technique. Bacterial enumeration of this organism followed the procedure outlined in Standard Methods[13] (see Appendix A).

Further identification of Pseudomonas spp. and Pseudomonas-like bacteria was performed with reference to the scheme of Shayegani et. al.[18], and enteric bacteria were identified by using the tests listed in Figure 9[17], Manual of Clinical Microbiology[19], and Biochemical Tests for Identification of Medical Bacteria[20].

Enumeration of sulfate-reducing bacteria--The enumeration of sulfate-reducing bacteria involved the use of several methods. The media used are listed in Appendix A.

Initially, qualitative measurement of sulfate-reducers in 24-hour standing pipe loop water was carried out using Baars' medium supplemented with yeast extract and reducing agents. The culture medium added to a 60 mL bottle containing 30 mL of sample. The bottle was filled to the brim and stoppered to exclude air. Incubation was at 28-30°C for 4-21 days. After growth was indicated by the appearance of black precipitates (iron sulfide), a cell suspension from the clear supernatant in the upper portion of the medium was centrifuged and washed twice in 3% peptone water (pH 7.2). This culture was then streaked on Lewis medium solidified with 1.5% agar; incubated at 28-30°C in an anaerobic jar with H_2 (BBL GasPak system). The pour plate method was also employed using the Lewis medium and incubation was at 28-30°C in an anaerobic jar with H_2. Growth was indicated by black colonies.

For quantitative measurement of sulfate-reducers in pipe loop water, the following procedure was used. An MPN was obtained after five sterile screw cap test tubes were inoculated with 1 mL each of the 24-hour standing pipe loop water, five 16 x 150 mm test tubes were each inoculated with 10 mL of the standing water, and five 16 x 150 mm test tubes were each inoculated with a 0.45 µm membrane filter through which 100 mL of standing water had been filtered. Sterile medium was then poured to the 5 cm mark at the top of each tube. The tubes were closed immediately by sterile rubber stoppers and the contents were mixed by inversion 6 times. Following this step, a 4-5 cm plug of sterile 1.5% agar was poured in and the tube closed by a screw cap. Tubes with sulfate-reducers demonstrated blackening within 21 days of incubation at 28-30°C.

Quantitative enumeration of sulfate-reducers in pipe incrustations, both before and after corrosion treatment, was performed using the Lewis medium and a 5-tube MPN series. For inoculation, the tubercles were extracted from the pipe with a sterile scalpel, manually ground in a sterile mortar, weighed aseptically, and diluted 1:10 with sterile dilutent. Decimal dilutions were made to provide a MPN using the Lewis medium.[21] Tubes with sulfate-reducers showed blackening with 21 days of incubation at 28-30°C.

Enumeration of iron-oxidizing bacteria--Qualitative analysis of 24-hour standing pipe loop water for *Gallionella* was performed using ferrous sulfide agar (Appendix A).

Ferrous sulfide agar slants[22] were prepared and the tubes inoculated with 1- and 5-mL portions of the standing water. Growth at ambient temperatures usually occurred in 18-36 hours and appeared as a white deposit on the sides of the test tube. Growth of *Gallionella* was also received with use of ferrous sulfide agar supplemented with formalin.[23]

Microscopic observation of *Gallionella* was facilitated by the Prussion blue reaction.[13, 22] Cell suspensions from the cultures were carefully placed on slides and allowed to dry. Several drops of 1 N HCl were added to the dried colonies followed by a drop of a 1% solution of potassium ferrocyanide. A blue precipitate of Prussion blue was formed as the iron around the cells or filaments was dissolved.

Liquid broth B_{12} medium (Appendix A) was employed to culture the following iron-oxidizing bacteria: *Gallionella*, *Sphaerotilus*, and *Caulobacter*. Qualitative analysis of these organisms in 24-hour standing pipe loop water consisted of inoculating sterile screw cap test tubes (16 x 150 mm) containing 10 mL of B_{12} broth each with a 0.45 µm membrane filter through which 100 mL of standing water had been filtered. After incubation at 24°C for 48-72 hours, growth was exhibited as a mucous film attached to the sides of the test tube and near the surface of the media. The liquid broth was cloudy with small visible particles floating in it.

Headloss

Headloss in the test pipe loop system was determined from water pressure and flow velocity measurements and the application of the Bernoulli equation. Water pressure measurements were made with pressure test gauges located at the beginning and end of the pipe loop system.

Scanning Electron Microscopy

Preparation of specimens for scanning electron microscopy (SEM) and X-ray energy dispersive microanalysis generally was as follows:

Water Samples: Standing, Running and Control samples: 20, 50 and 100 mL volumes of each sample were filtered through 0.2 µm Nuclepore filters; specimens were then fixed in an ice-cold solution of 2.5% (wt/vol) electron microscopy-grade glutaraldehyde in 50 mM monobasic potassium phosphate buffer, pH 7.0 for 30 minutes at 23°C. After fixation, the specimens were gently washed twice in sterile prefiltered distilled water; dehydrated in an increasing ethanol concentration series (30, 50, 75, 85, 95, and 100% x 3), and transferred to a Freon 113 series before critical point drying (CPD) according to Cohen _et_. _al_.[24] The specimens were then mounted on aluminum stubs with double stick tape and rendered electrically conductive by coating with vaporized carbon and 200-300 Å Au/Pd.

Tubercles: Tubercles were carefully extracted from the pipe surface with a sterilized scalpel and fixed in an ice-cold solution of 2.5% (wt/vol) electron microscopy-grade glutaraldehyde in 50 mM monobasic potassium phosphate buffer, pH 7.0. After overnight fixation at 4°C, the tubercles were prepared in the same manner as the water samples; however, they were mounted to the aluminum stubs with silver collodial adhesive rather than tape. On some of the tubercles, partial dissolution of amorphous ferric iron was achieved by acidifying the samples with 1% and 5% solutions of oxalic acid for 30-45 minutes prior to the preparation for electron microscopy.[25]

SECTION 6

RESULTS AND DISCUSSION

ELECTROCHEMICAL TESTS

General

The electrochemical tests were designed to measure the effects of chlorination on the corrosion rate of copper in Cedar River water. Data was collected for chlorine residuals dosed as NaOCl and Cl_2. When chlorine gas Cl_2 is added to water, hypochlorous acid, HOCl, is formed rapidly as the chlorine is hydrolyzed.[26]

$$Cl_2 + H_2O \rightarrow H^+ + Cl^- + HOCl$$

Hypochlorous acid is also formed when sodium hypochlorite, NaOCl, is added.

$$NaOCl + H^+ \rightleftharpoons Na^+ + HOCl$$

Depending on solution pH, hypochlorite ion OCl^- may be present in appreciable quantities.

$$HOCl \rightleftharpoons H^+ + OCl^-$$

Hypochlorous acid is the most important species for disinfection and a powerful oxidizing agent which can be reduced to water and chloride ion.

$$H^+ + Cl^- + HOCl + 2e^- \rightleftharpoons H_2O + 2Cl^-$$

Whether added as chlorine gas or sodium hypochlorite, chlorination tends to alter the pH, increase the chloride concentration, and add a chlorine residual to the water. The effect of each of the changes on corrosion rate was measured; pH was varied from approximately pH 6 to pH 8, chloride concentration was varied from 3.0 to 11.0 mg/L and free chlorine residual was varied from 0 to 2.0 mg/L. The effects of chloride concentrations and chlorine residual were measured at both pH 7.0 and pH 8.0. Data was collected for chlorine residuals dosed as NaOCl and Cl_2.

Electrode Preparation

Copper electrodes aged for several weeks in Cedar River water with an appreciable chlorine residual (>0.2 mg/L) were found to be highly pitted and not protected by the continuous oxide film that grows naturally on copper in unchlorinated Cedar River water. Pitting made it effectively impossible to measure corrosion rates electrochemically on some specimens. It was found that corrosion rates could be measured with some certainty on freshly polished copper electrodes so the bulk of the corrosion rate measurements were made on freshly polished electrodes. Even on freshly polished, oxide-free surfaces, the effects of localized corrosion could be seen after each polarization test was run. The severity of localized attack tended to increase with increasing chlorine residual. Reproducibility of corrosion rate measurements was diminished as localized attack became more pronounced.

All data was taken using the rotating disc electrode at a rotation speed of 3000 RPM. At this rotation speed, any reduction in corrosion rate due to solution mass transport of reacting species was effectively eliminated. Thus, corrosion rates measured under these conditions were limited only by the kinetics of the corrosion reaction.

The magnitudes of kinetically measured corrosion rates are considerably higher than those measured under the combined effects of kinetics and solution mass transport. The effects of pH, chloride concentration, and free chlorine residual shown in the figures that follow are therefore exaggerated as compared to the effects that are actually manifested in a corroding system. This exaggeration allows for a clear representation of the qualitative effects of each parameter on the overall corrosion rate. It also provides a relative basis for comparing the impact of variations in pH, chloride concentration, and free chlorine residual on the corrosion rate of copper in Cedar River water.

The Effect of pH on Corrosion Rate

The corrosion rate of copper in Cedar River water was measured at pH values from 6.0 to 8.0. In each test the pH was adjusted by the addition of either $CO_2(g)$ or 0.1 M CaO. Only slight amounts of each were needed to overcome the very low buffer capacity of Cedar River water and alter the pH. Figure 10 shows the effects of pH on corrosion rate at 25°C in unchlorinated Cedar River water. These results are similar to previous pilot plant test results on Cedar and Tolt water [2, 3] and with other electrochemical test results on synthetic Tolt water.[9] A reduction in corrosion rate of roughly 80% is achieved by raising the pH from 6.0 to 8.0, and a reduction of approximately 60% is possible by raising the pH from 7.0 to 8.0. Overall, the change in corrosion rate due to variation in solution pH is limited to a range of 1.6 to 0.1 mils per year.

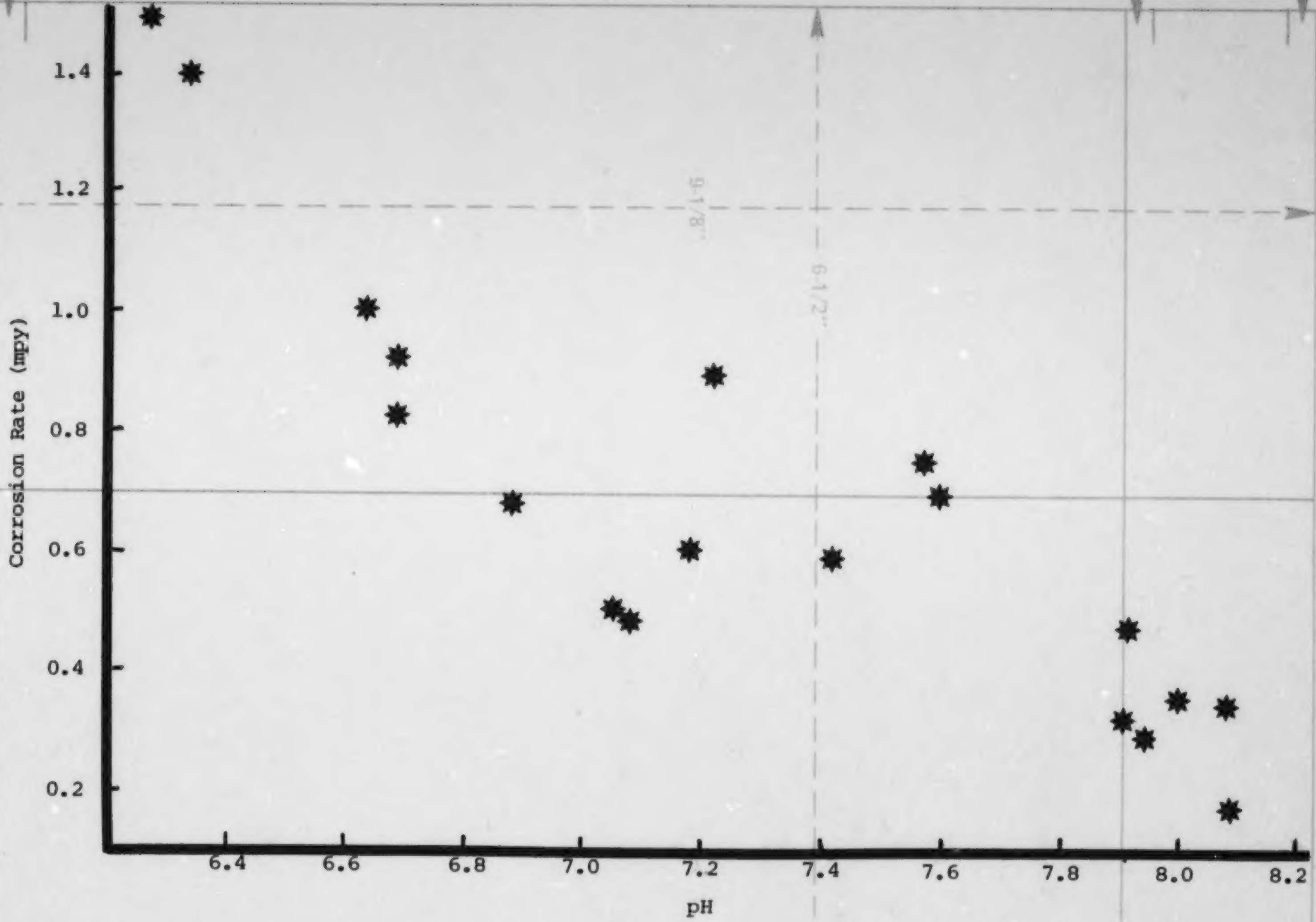


Figure 10. The effects of pH on copper corrosion rates in unchlorinated Cedar water.

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Effect of Chloride Ion on Copper Corrosion Rate

Chloride ion concentration was varied over a range of 3 to 11 mg/L at both pH 7 and 8. The rate of copper corrosion determined at these varied chloride concentrations and at 25°C is presented in Figure 11. Note the slight tendency for corrosion rate to increase with increasing chloride ion concentration, but overall the rate was limited to a range of 0.1 to 0.8 mils per year.

Effect of Free Chlorine Residual on Copper Corrosion Rate

Chlorine residual was varied from zero up to approximately 2 mg/L by addition of NaOCl and Cl_2. Data collected at both pH 7 and 8 are plotted in Figure 12. This plot shows a significant increase in the range of corrosion rate values measured over the small range of free chlorine residual variation. Overall, corrosion rates ranged from approximately 0.5 mpy to zero residual to roughly 12.5 mpy at a residual of 1.8 mg/L.

Comparison of Effects

Over the range of variation of pH values, chloride concentrations, and chlorine residuals tested in Cedar River water, chlorine residual was shown to have by far the most profound impact on corrosion rate. An order of magnitude comparison of the effects of each of these chemical variables is presented in Figure 13.

Effect of Raising the pH from 7.0 to 8.0 on Corrosion Rates at Varied Chlorine Residual Levels

To evaluate the effect of raising the pH from 7 to 8 on copper corrosion in the presence of a chlorine residual of varied magnitude, the data was plotted as corrosion rate versus pH for small ranges in free chlorine residual. Data taken at free chlorine residual values of 0 to 0.2 mg/L are plotted to show the effect of pH at that residual level. As shown in Figure 14, raising the pH from 7 to 8 reduces the corrosion rate by approximately 75%.

Data taken at free chlorine residual levels of 0.2 to 0.4 mg/L are plotted in Figure 15, and also demonstrate a reduction in corrosion rate of approximately 75%, raising the pH from 7 to 8.

Data taken at free chlorine residual levels above 0.4 mg/L are presented in Figure 16. The data scatter was considerable and overshadowed any possible detection of a trend.

An overview of the findings of this study on increasing chlorine residual over the pH range of 7 to 8 is shown in Figure 17. This data suggests that at free chlorine residual levels above 0.4 mg/L the effectiveness of raising the pH from 7.0 to 8.0, to reduce the corrosion rate of copper, may be lessened.

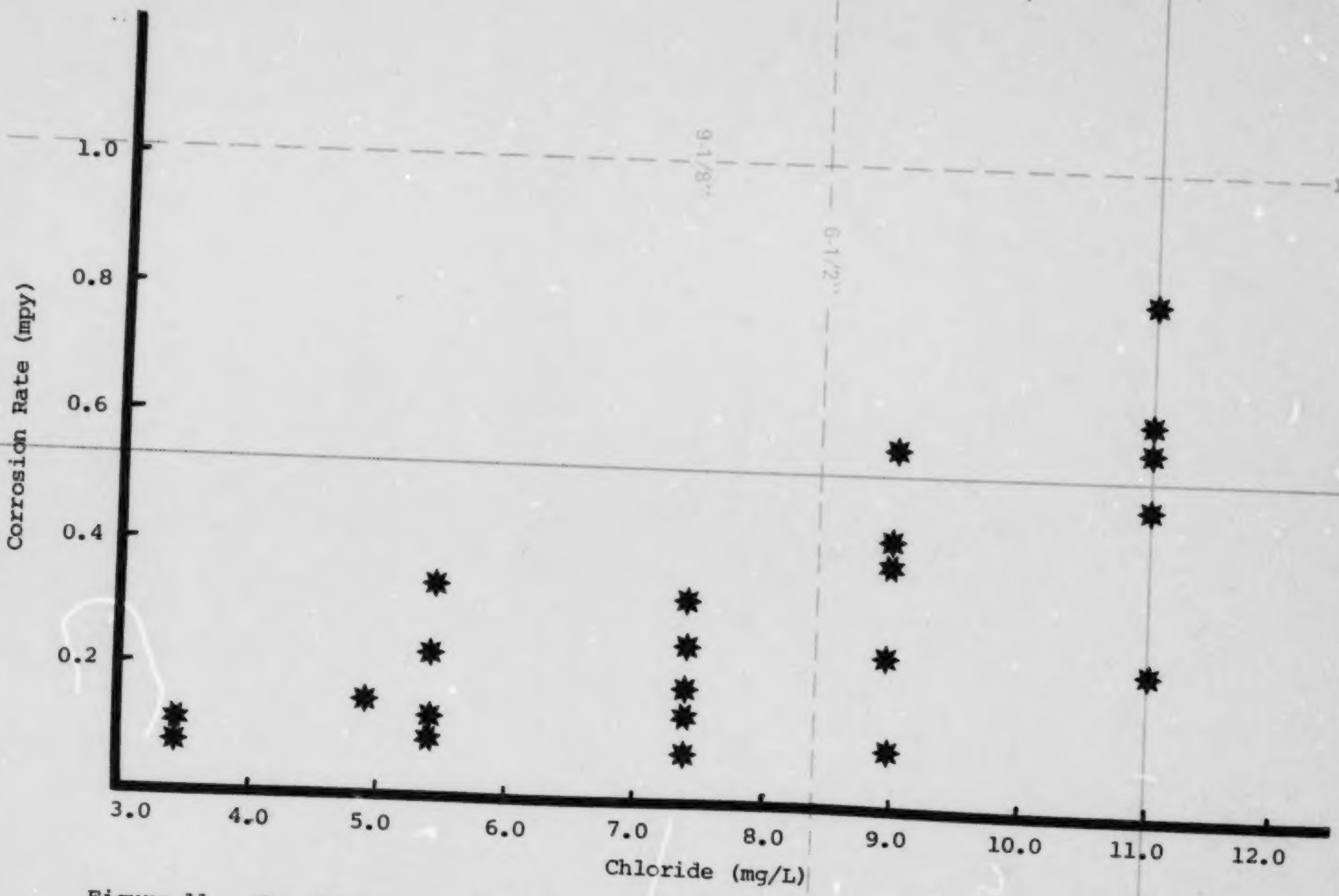

Figure 11. The effects of chloride on copper corrosion rates in unchlorinated Cedar water.

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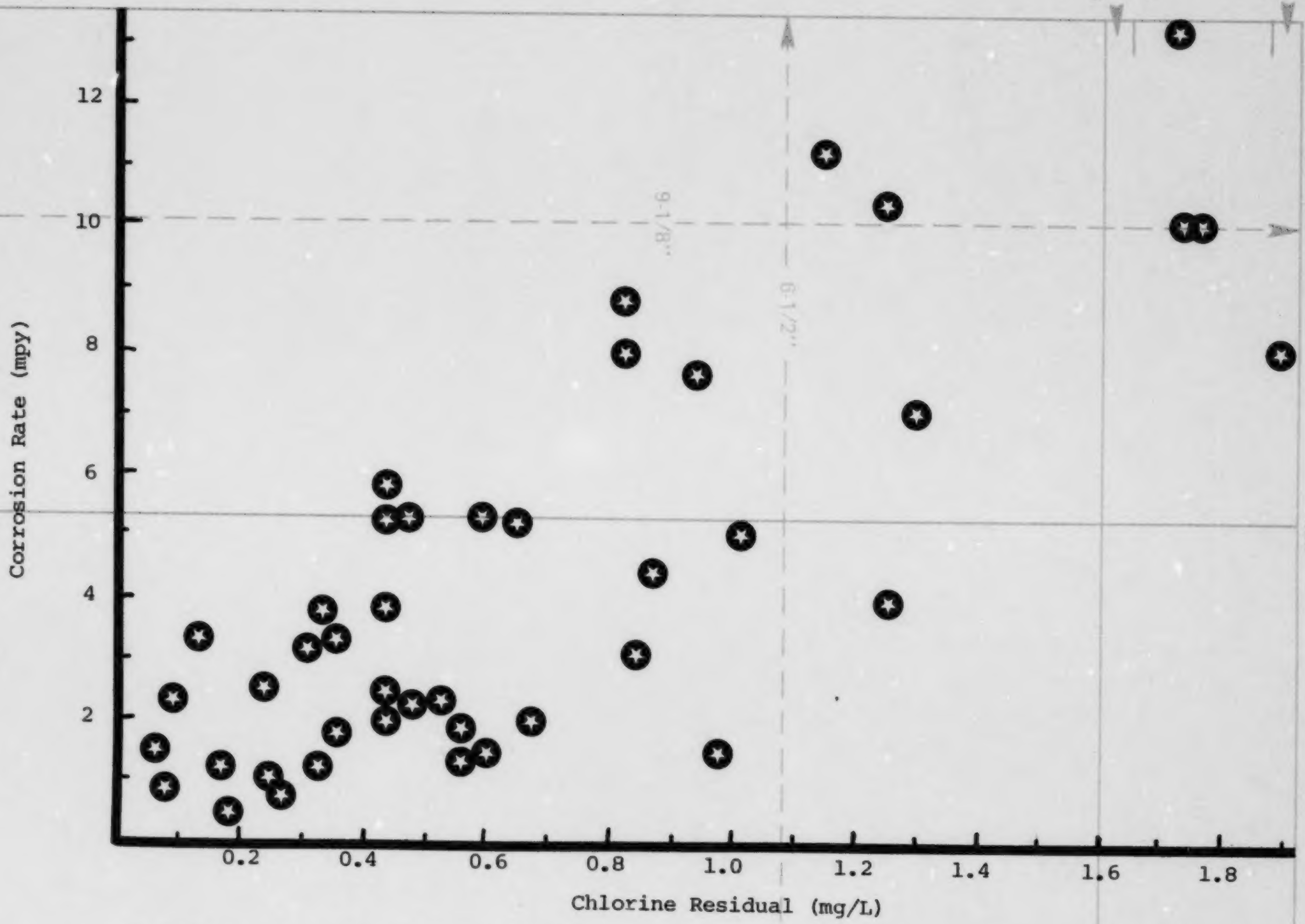

Figure 12. The effects of free chlorine residual on copper corrosion rates in Cedar water (dosed as NaOCl and Cl_2).

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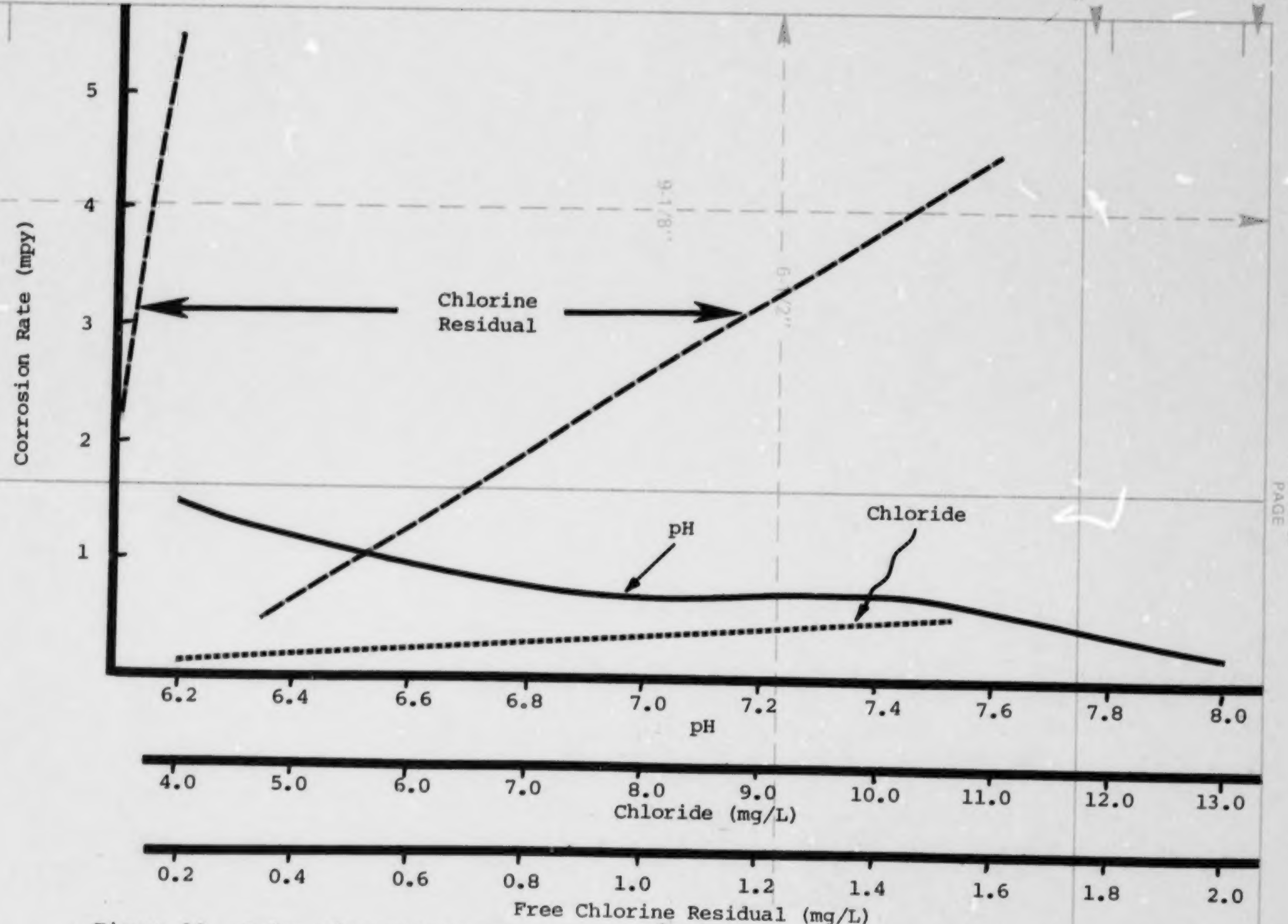

Figure 13. Order of magnitude comparison of the effects of pH, chloride, and chlorine residual on copper corrosion rates in Cedar water.

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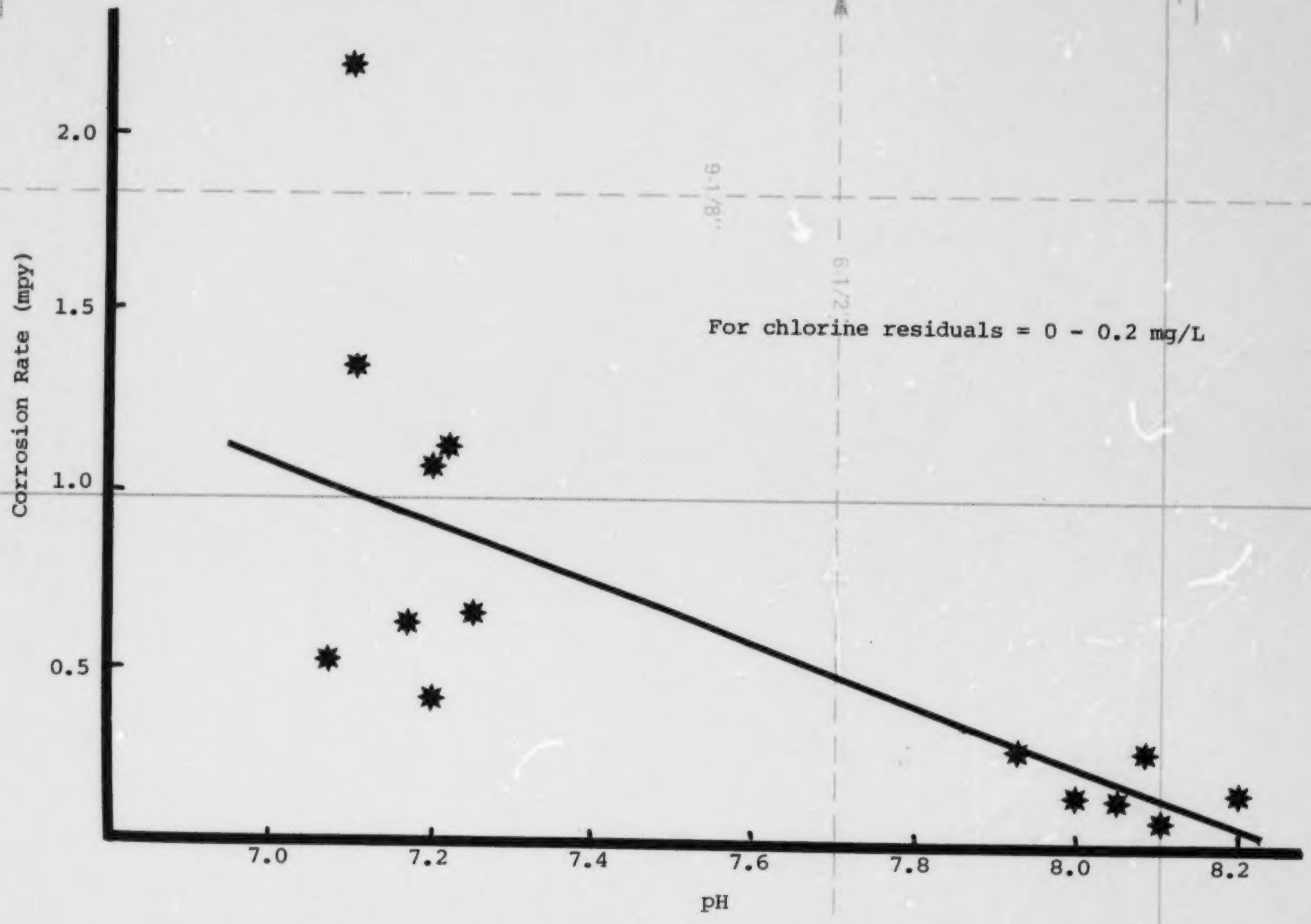

Figure 14. The effects of pH on copper corrosion rates in chlorinated Cedar water.

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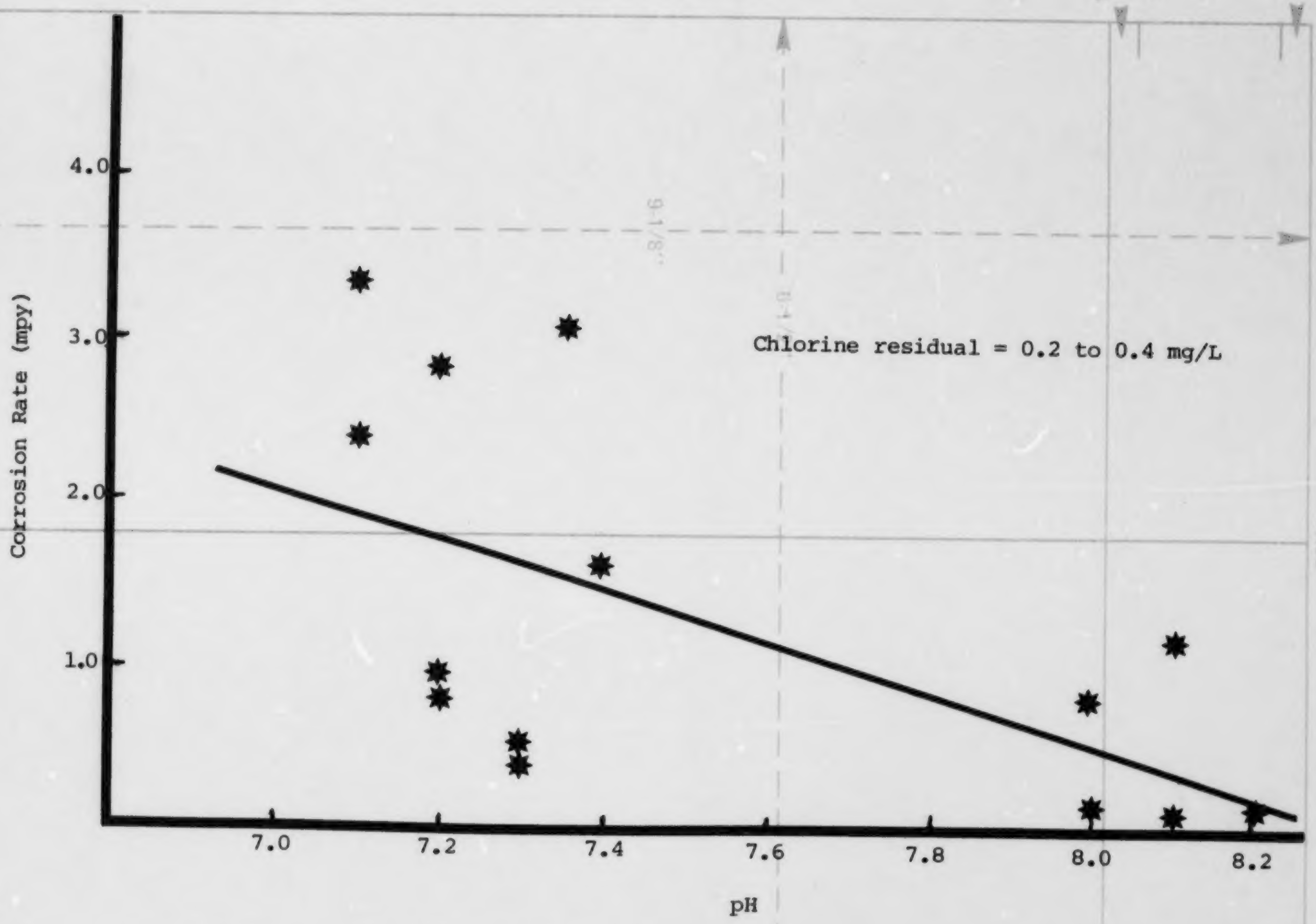

Figure 15. The effects of pH on copper corrosion rates in chlorinated Cedar water.

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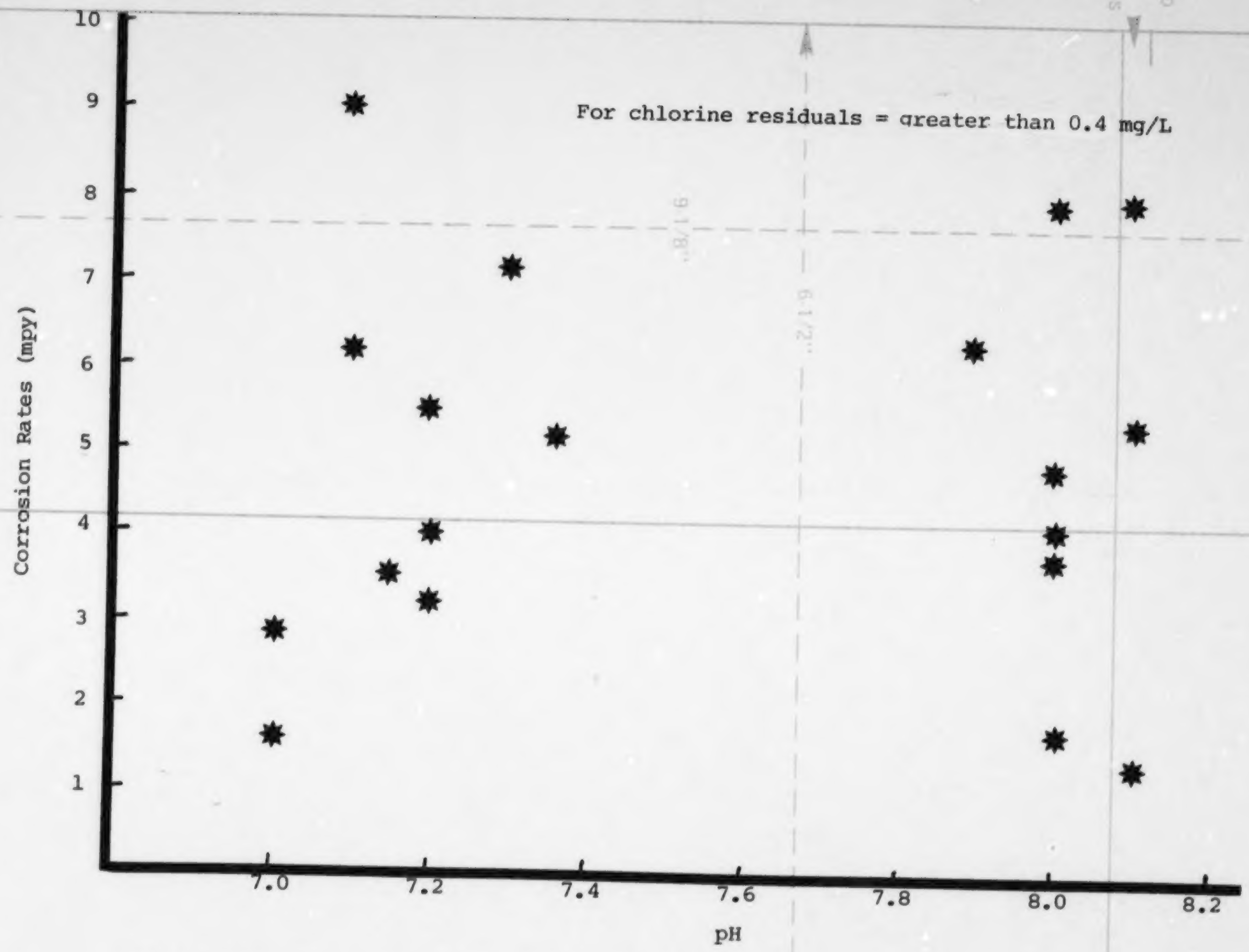

Figure 16. The effects of pH on copper corrosion rates in chlorinated Cedar water.

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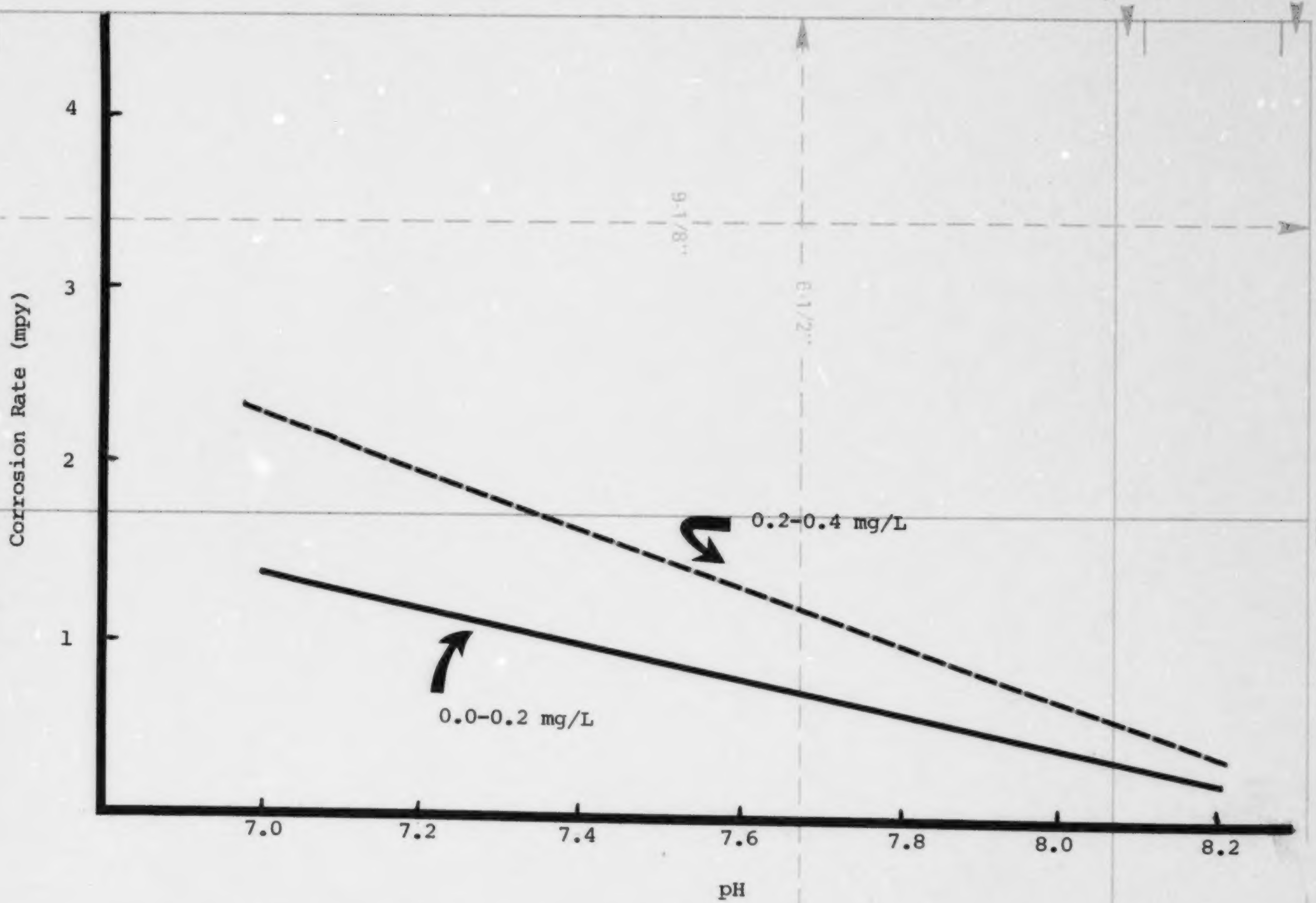

Figure 17. The effects of pH on copper corrosion rates in chlorinated Cedar water.

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Although not conclusive, these tests show that the chlorine residual level maintained in Cedar River water has a big impact on the rate of copper corrosion and suggest that a more detailed study of the problem is warranted.

Comparison of the Effects of NaOCl Dosed Chlorine Residuals and Cl_2 Dosed Residuals on Copper Corrosion at pH 7 and 8

Identical tests were run at pH 7 and 8 and varied chlorine residual levels for Cedar River water dosed with NaOCl and Cl_2. The data are presented as corrosion rate versus chlorine residual at a specified pH.

At pH 7, Figures 18 and 19 show the variation in corrosion rate with free chlorine residual dosed as Cl_2 and NaOCl, respectively. Both treatments are shown together in Figure 20, and approximately the same variation in corrosion rates occur for both treatments. At pH 8, the same variations occur once again for both treatments.

This data was taken realizing that adding a chlorine residual as Cl_2 tends to drive the pH down more than adding NaOCl. The pH effect was shown earlier and this comparison was conducted to observe any differences in corrosivity existing between the chlorine residuals dosed as NaOCl or Cl_2. The data presented here suggest that approximately equal corrosion rates occur with chlorine residuals dosed as NaOCl and Cl_2.

Summary of the Electrochemical Tests

The electrochemical tests demonstrated that chlorination using both NaOCl and Cl_2 greatly increases corrosion rates in copper plumbing systems. Although substantial corrosion reduction can be achieved by pH adjustment to pH 8 at a given chlorine dosage, the benefits achieved by the pH adjustment would be diminished if chlorine dosages were increased above 0.4 mg/L.

MICROBIOLOGICAL LABORATORY TESTS

General

The impact of corrosion treatment on chlorine disinfection was determined as described in Section 5. Specifically, these tests were conducted to determine if SWD would be required to increase chlorine dosages for adequate disinfection when corrosion treatment is implemented. In these tests, three variables involved in the disinfection process (pH, free chlorine residual, and contact time) were varied and the effects of each variable are discussed below.

Effect of pH on Disinfection

The importance of pH in chlorine disinfection was previously discussed in Section 1. Other things being equal, the germicidal efficiency of free chlorine residual is a function of the pH, which establishes the amount of dissociation of HOCl to H^+ ion and the OCl^- ion. The OCl^- ion is a relatively poor disinfectant because of its inability to diffuse through the

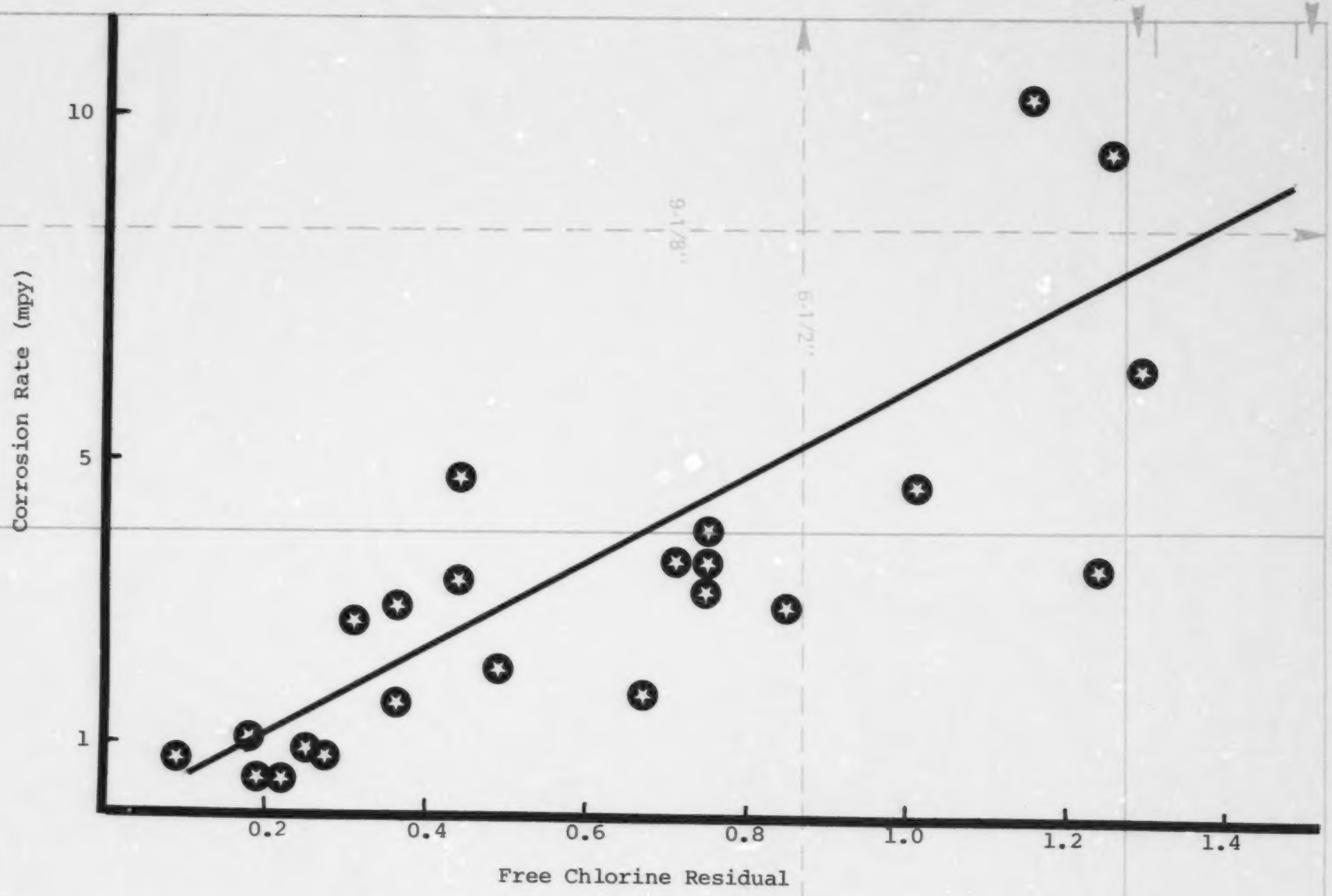

Figure 18. The effects of chlorine residual dosed as Cl_2 on copper corrosion rates in pH 7 Cedar water.

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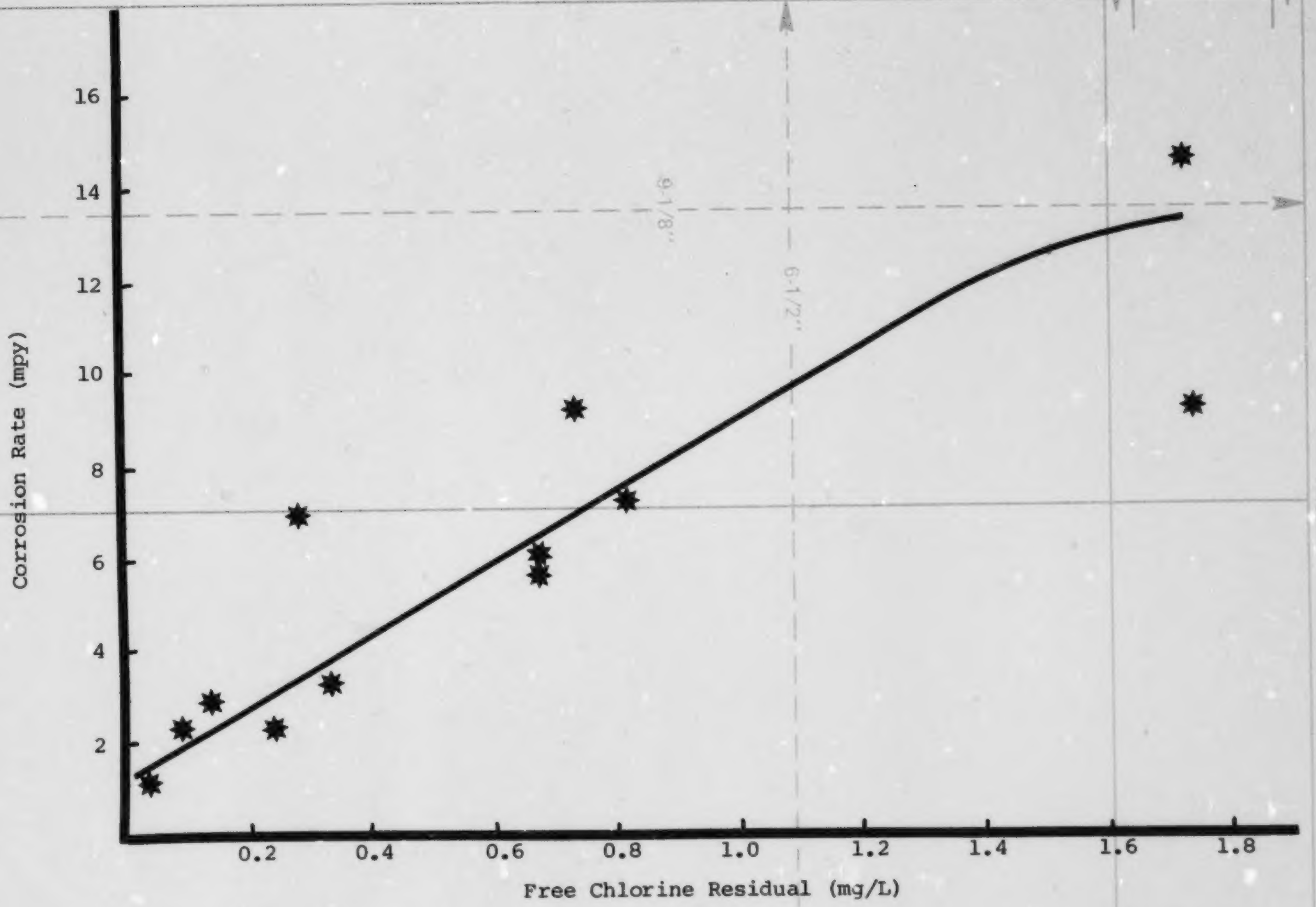

Figure 19. The effects of chlorine residual dosed as NaOCl on copper corrosion rates in pH 7 Cedar water.

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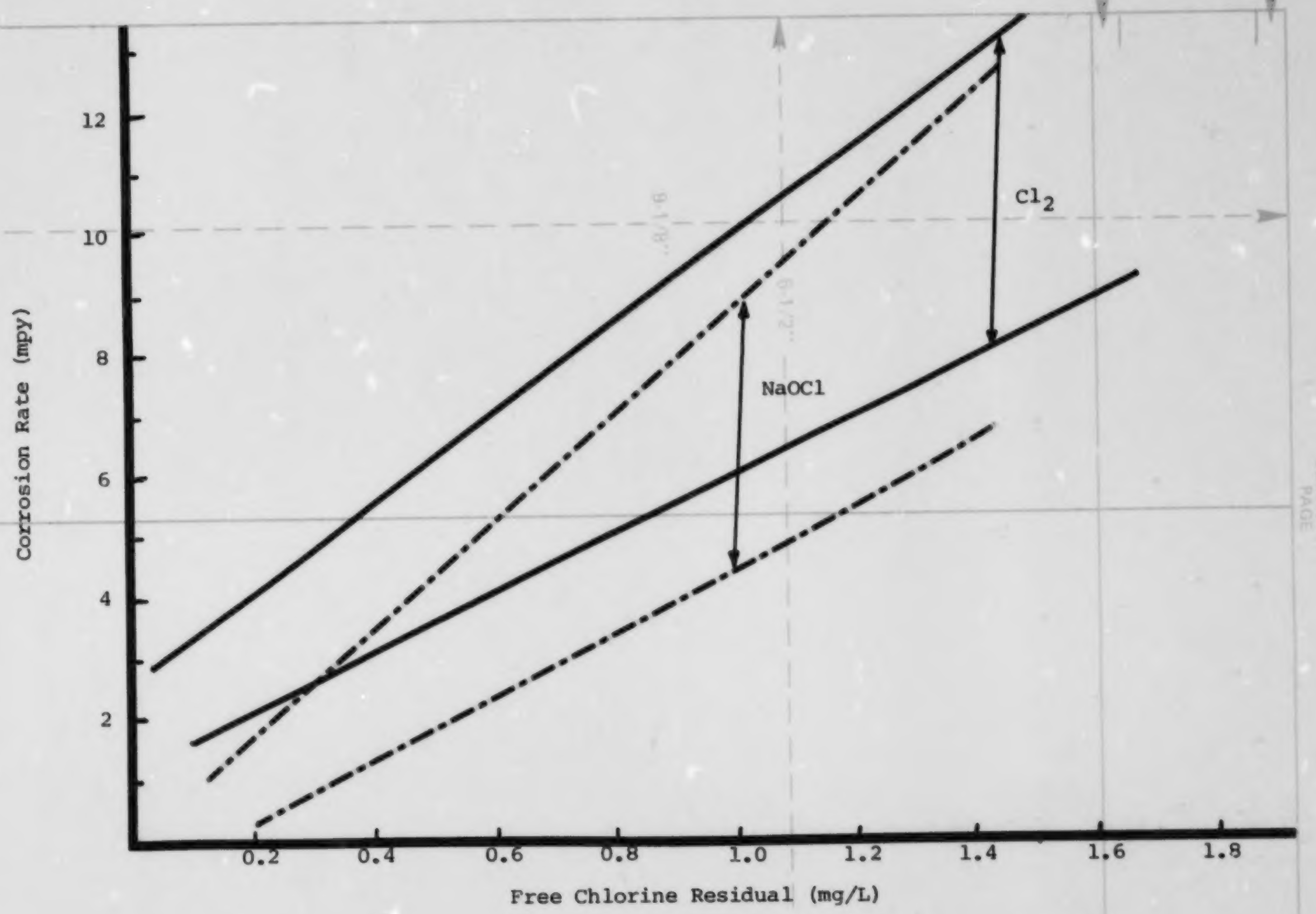

Figure 20. Comparison of the effects of chlorine residual dosed as Cl_2 and NaOCl on copper corrosion rates in pH 7 Cedar water.

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cell wall of microorganisms.[26]

From Table 6 it is evident that, in these tests, the amount of HOCl present at pH 8 was much less than that present at pH 6 or 7. Table 6 demonstrates that since HOCl is far more bacteriocidal than OCl^-, a slower death rate occurred at pH 8 than at pH 6 or 7. At pH 8, the HOCl concentration for the free chlorine residual tested varied over a small range (0.002-0.006), whereas at pH 6 the HOCl concentration varied over a wider range (0.01-0.36), with the highest kill rate occurring at the highest HOCl concentration. Thus, an overall pH effect was evident in the kill rates with the lower concentrations of HOCl at pH 8 producing the lower rates of kill.

In Figures 21 through 29, the pH effect is readily apparent in the shape of the curves. At a given chlorine concentration, the curves for pH 7 (Cedar water) show a high initial rate of kill, i.e., 98-100% within the first 30 seconds after chlorine dosing. The percent bacteria killed over time for pH 6 (Tolt water) (Tables 7 through 9) also showed a high initial rate of kill which decreased over time. This decline in kill rate over time was probably due to a decreasing concentration of HOCl or the existence of small clumps of bacteria. Bacterial concentrations of the three different bacteria studied declined at different rates for pH 8, showing greater variability than when observed at pH 6 or 7 (Tables 7 through 9). The results also showed that the kill rates for Enterobacter aerogenes were much slower than those for either Pseudomonas aeruginosa or Escherichia coli. The kill rates for Enterobacter aerogenes were low initially and then increased as compared to the other two bacteria.

Many factors are involved in disinfection kinetics and it is the interaction of these factors which determines kill rates. Some of the factors are the rate of chlorine diffusion, lag time, accumulation rate, clumping of bacteria, variable cell resistances, and disinfectant species distribution.[27, 28] A high death rate initially is probably due to a rapid chlorine diffusion rate, low resistance of cells, and the high oxidizing ability of chlorine.[29] A declining death rate is usually due to a decrease in the concentration of HOCl and/or the presence of small or large bacterial clumps.[28] Sometimes death rates may decline because of poor disinfectant distribution or individual variability in cell resistance. A slow initial rate could also be due to lag time in accumulation of chlorine in cells. All of these factors may have been involved in these tests. Another possibility is the two-step death rate for bacteria postulated by Popp[30], where a high rate in the first 12 seconds is followed by a lower death rate.

Effect of Free Chlorine Residual on Disinfection

One would expect to see a lower concentration of bacteria at a given time as the chlorine concentration is increased, and the results from this study support this supposition. For pH 6, 7, and 8, the data (Tables 7-9) show that as the chlorine concentration was increased, the concentration of bacteria decreased. Results do not show much variability between the different pH levels in regard to this phenomenon.

TABLE 6. PERCENT KILL OF *P. AERUGINOSA*, *E. COLI*, AND *E. AEROGENES* AT VARIOUS CHLORINE CONCENTRATIONS

Organism	Free Residual Chlorine mg/L after 10 min.	HOCl Conc. mg/L	Average pH	Percent Kill		
				0.5 min.	1.0 min.	2.0 min
P. aeruginosa	0.01	0.01	6.48	79	79	79
	0.12	0.12	"	99	99	100
	0.28	0.27	"	100	100	100
E. coli	0.02	0.02	6.48	88	89	89
	0.23	0.22	"	98	100	100
	0.37	0.36	"	100	100	100
E. aerogenes	0.00	0.00	6.49	85	97	100
	0.11	0.11	"	95	100	100
	0.21	0.20	"	100	100	100
P. aeruginosa	0.01	0.01	7.09	99	100	100
	0.24	0.18	"	99	100	100
	0.34	0.26	"	100	100	100
E. coli	0.00	0.00	7.25	100	100	100
	0.10	0.08	"	98	100	100
	0.15	0.11	"	100	100	100
E. aerogenes	0.02	0.015	7.20	99	100	100
	0.13	0.10	"	100	100	100
	0.22	0.16	"	100	100	100
P. aeruginosa	0.01	0.002	8.03	67	68	68
	0.15	0.04	"	99	100	100
	0.25	0.06	"	100	100	100
E. coli	0.01	0.002	8.03	85	86	86
	0.17	0.04	"	98	100	100
	0.21	0.05	"	100	100	100
E. aerogenes	0.00	0.00	8.12	72	88	90
	0.13	0.03	"	86	99	100
	0.19	0.04	"	89	99	100

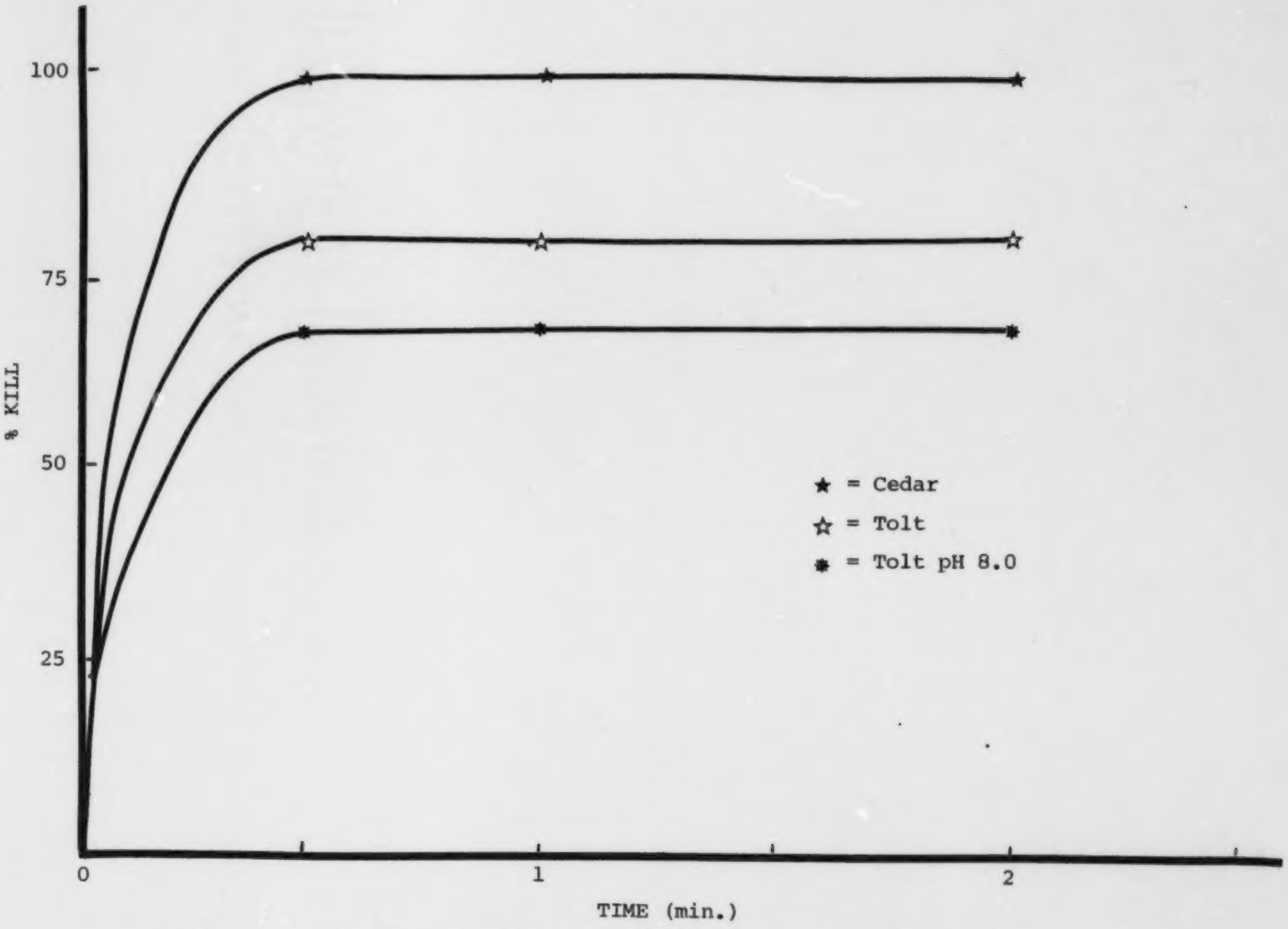

Figure 21. Percent kill of Pseudomonas aeruginosa at 0.1 mg/L chlorine dosage.

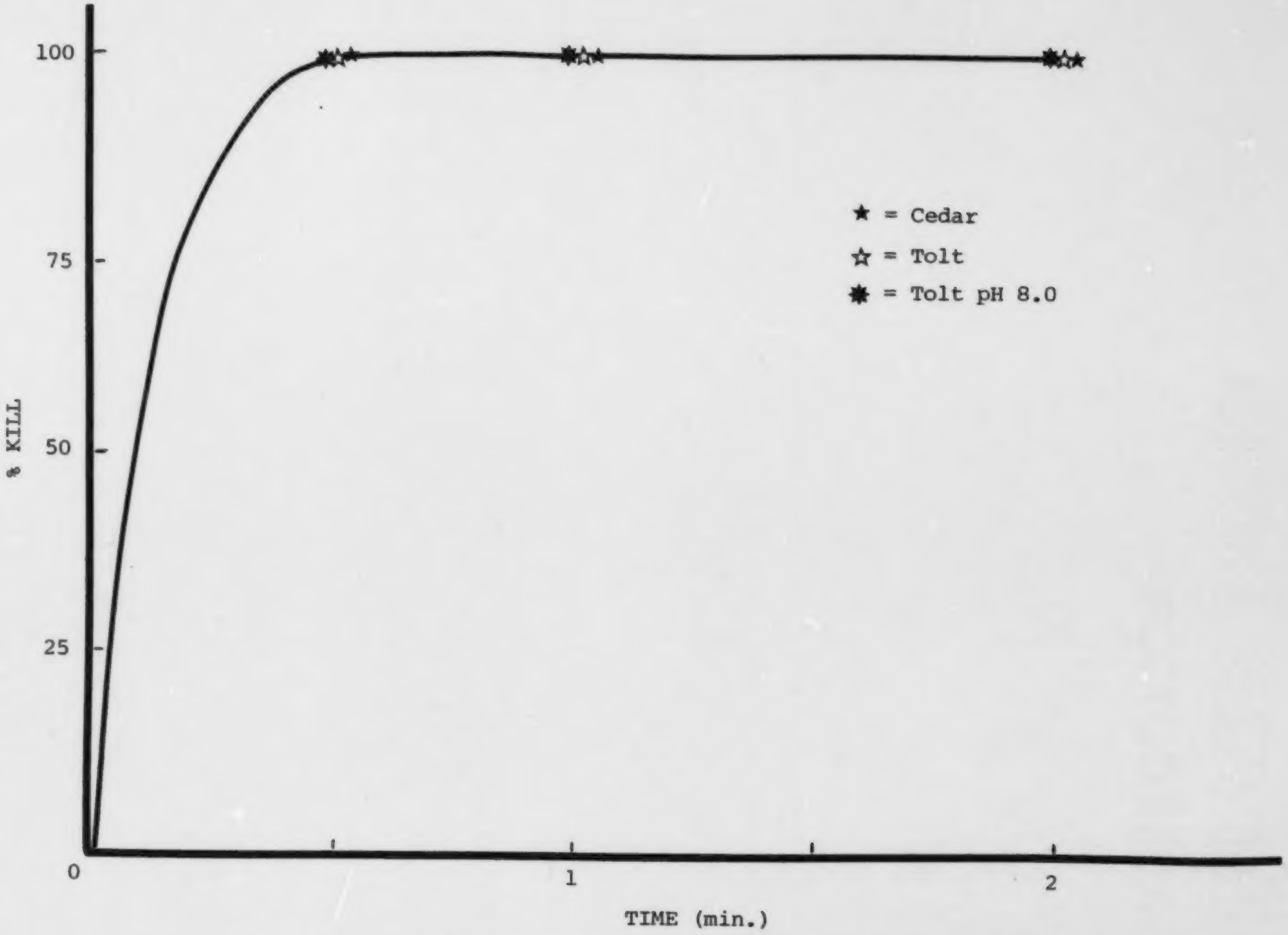

Figure 22. Percent kill of Pseudomonas aeruginosa at $[Cl_2] = 0.1$ mg/L after 10 minutes.

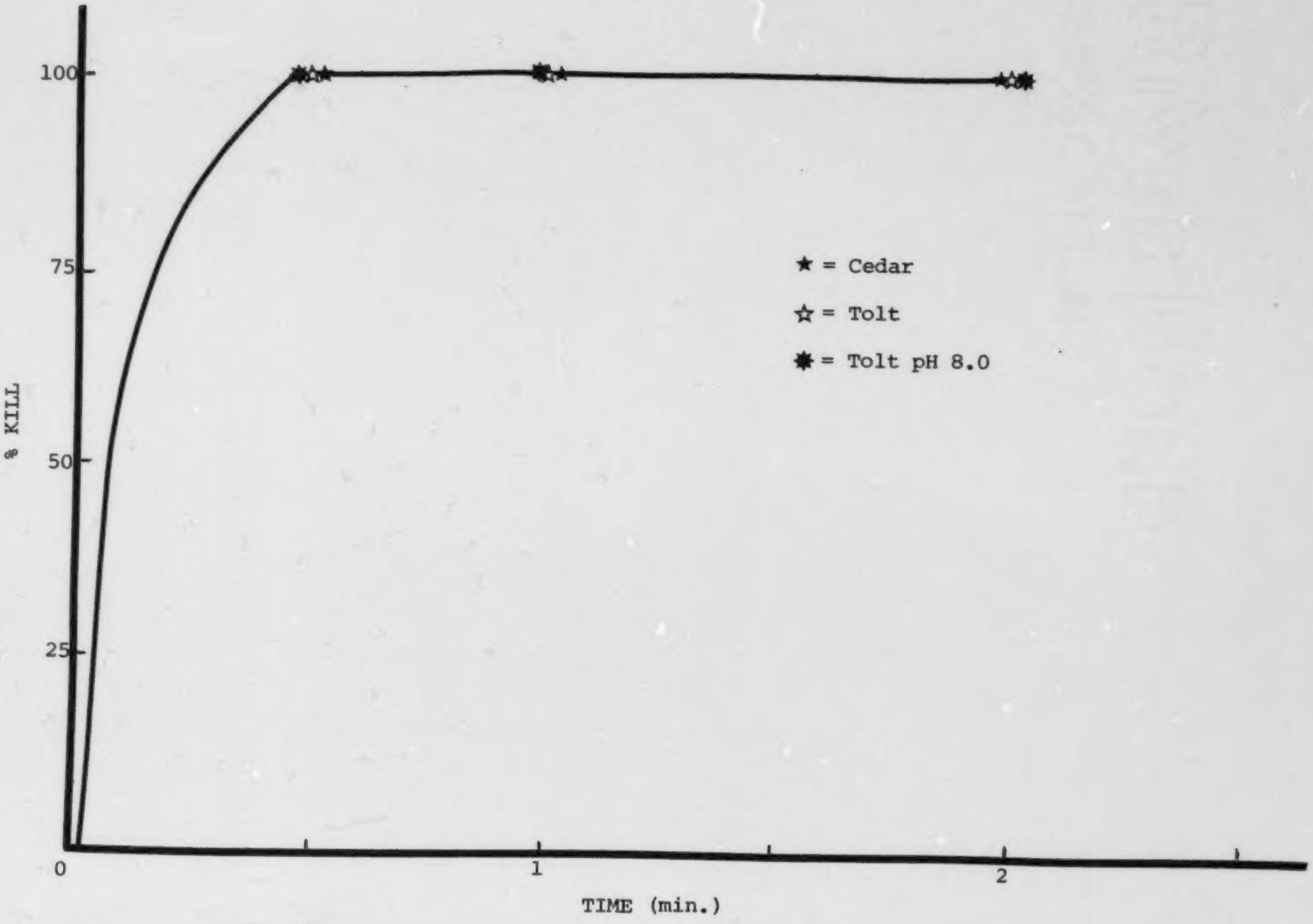

Figure 23. Percent kill of Pseudomonas aeruginosa at $[Cl_2]$ = 0.2 mg/L after 10 minutes.

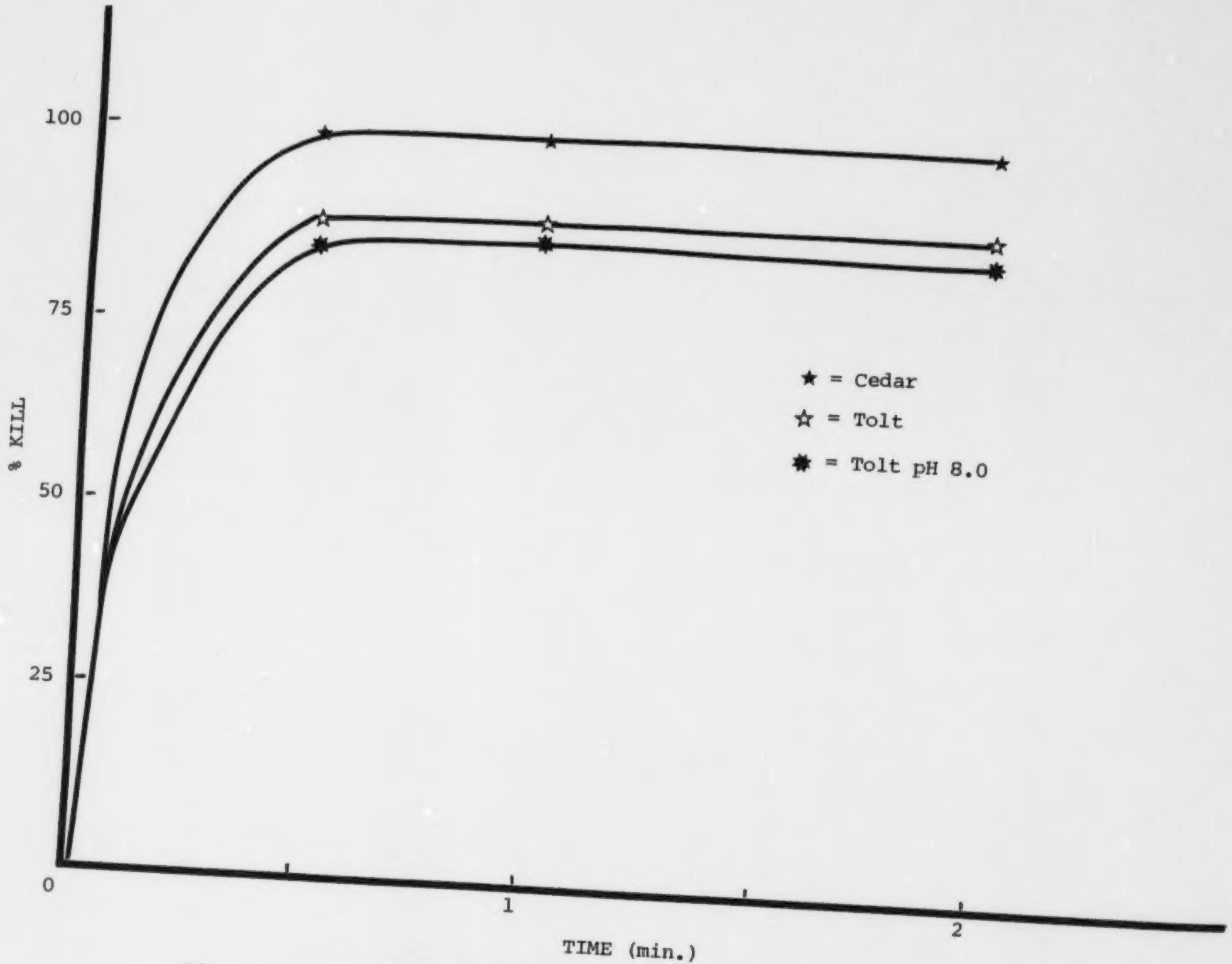

Figure 24. Percent kill of Escherichia coli at 0.1 mg/L chlorine dosage.

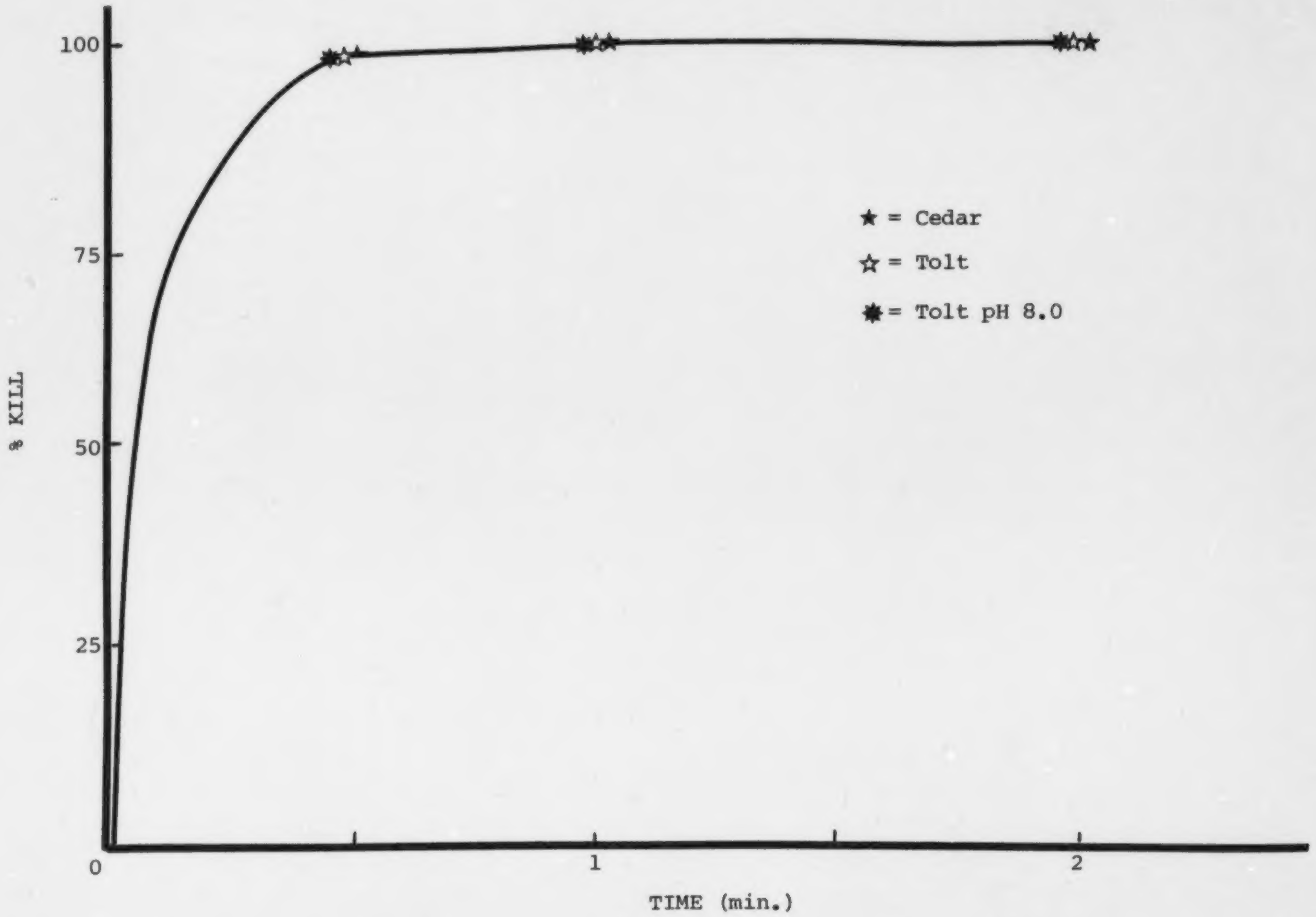

Figure 25. Percent kill of *Escherichia coli* at $[Cl_2]$ = 0.1 mg/L after 10 minutes.

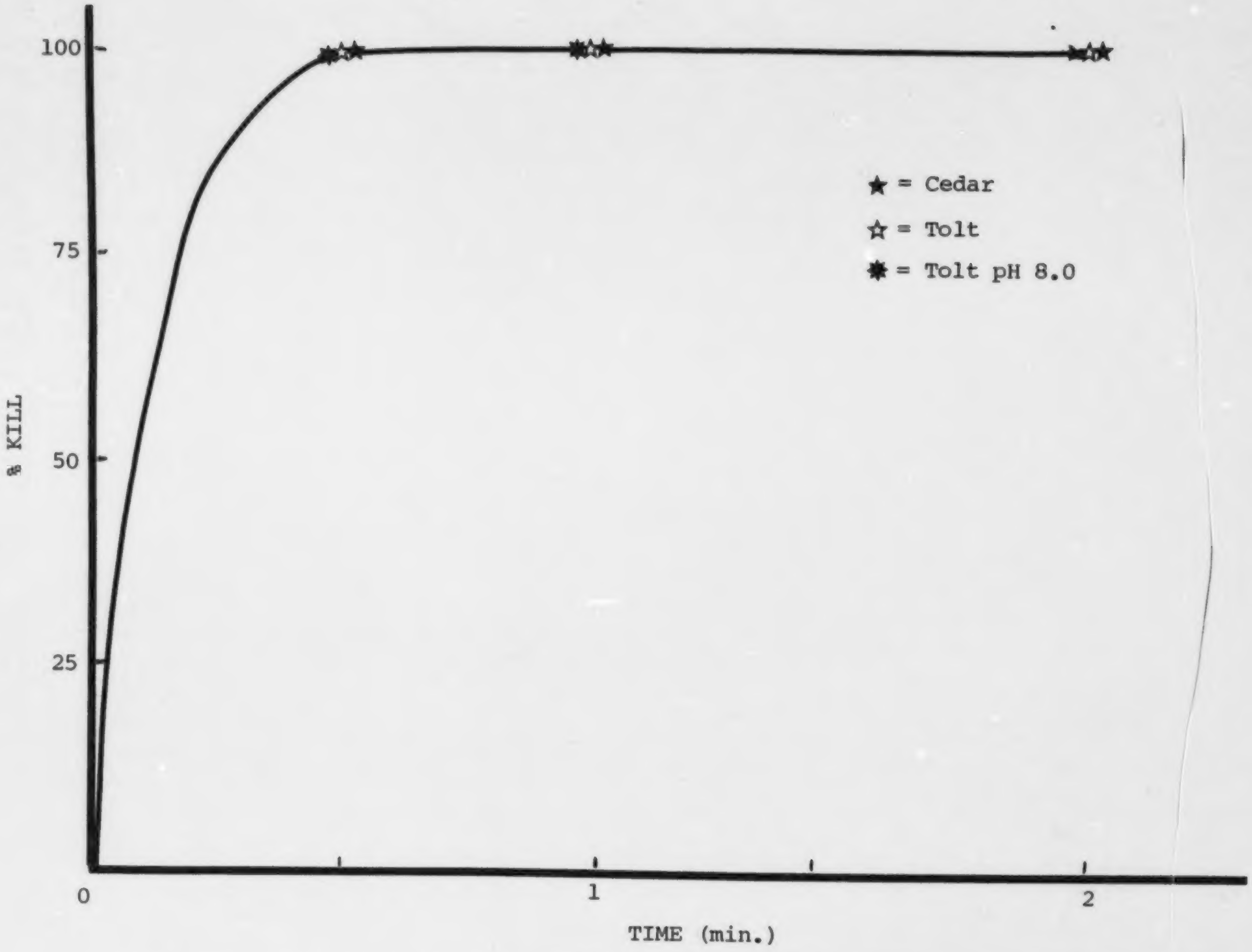

Figure 26. Percent kill of Escherichia coli at $[Cl_2]$ = 0.2 mg/L after 10 minutes.

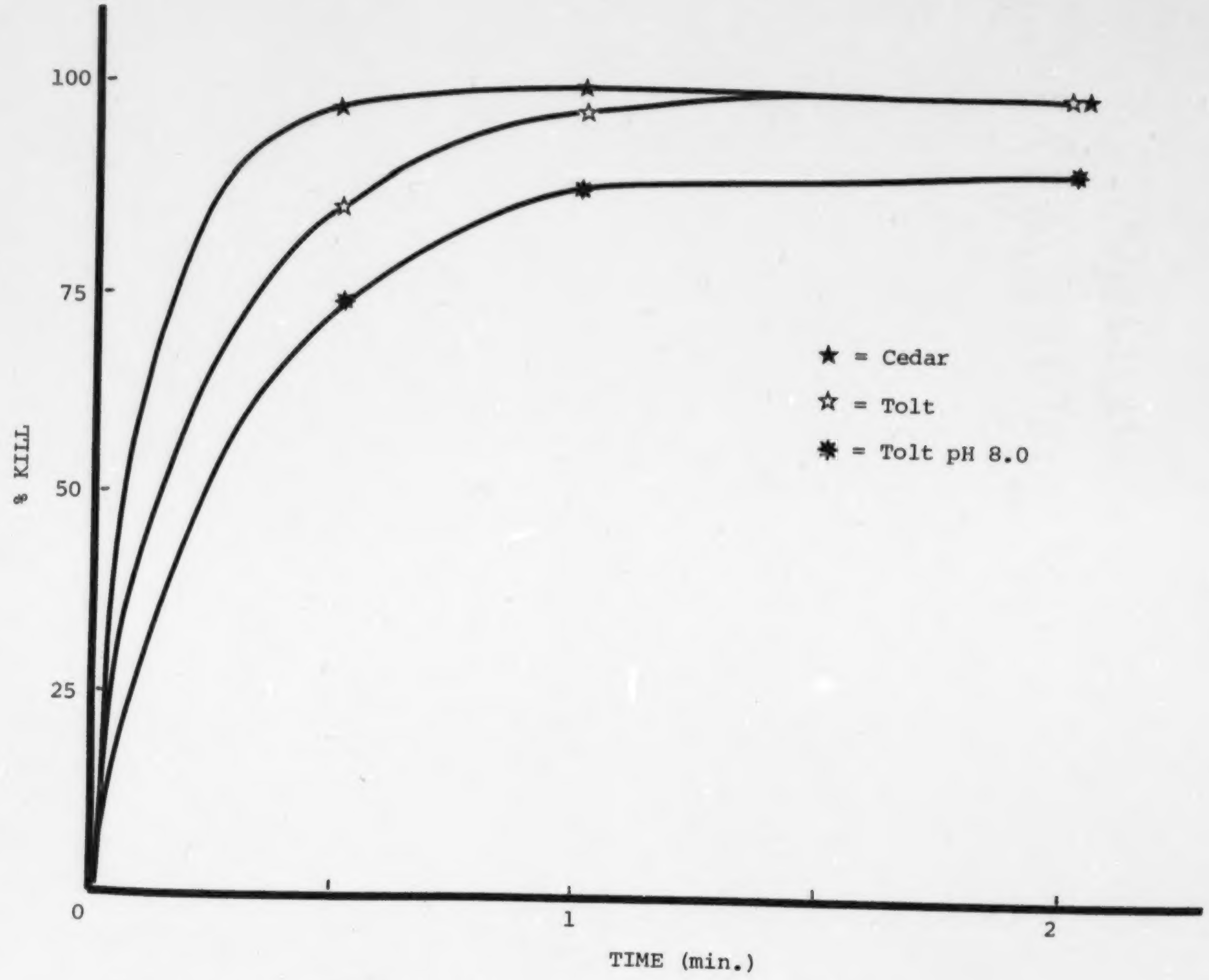

Figure 30. Percent kill of *Enterobacter aerogenes* at 0.1 mg/L chlorine dosage.

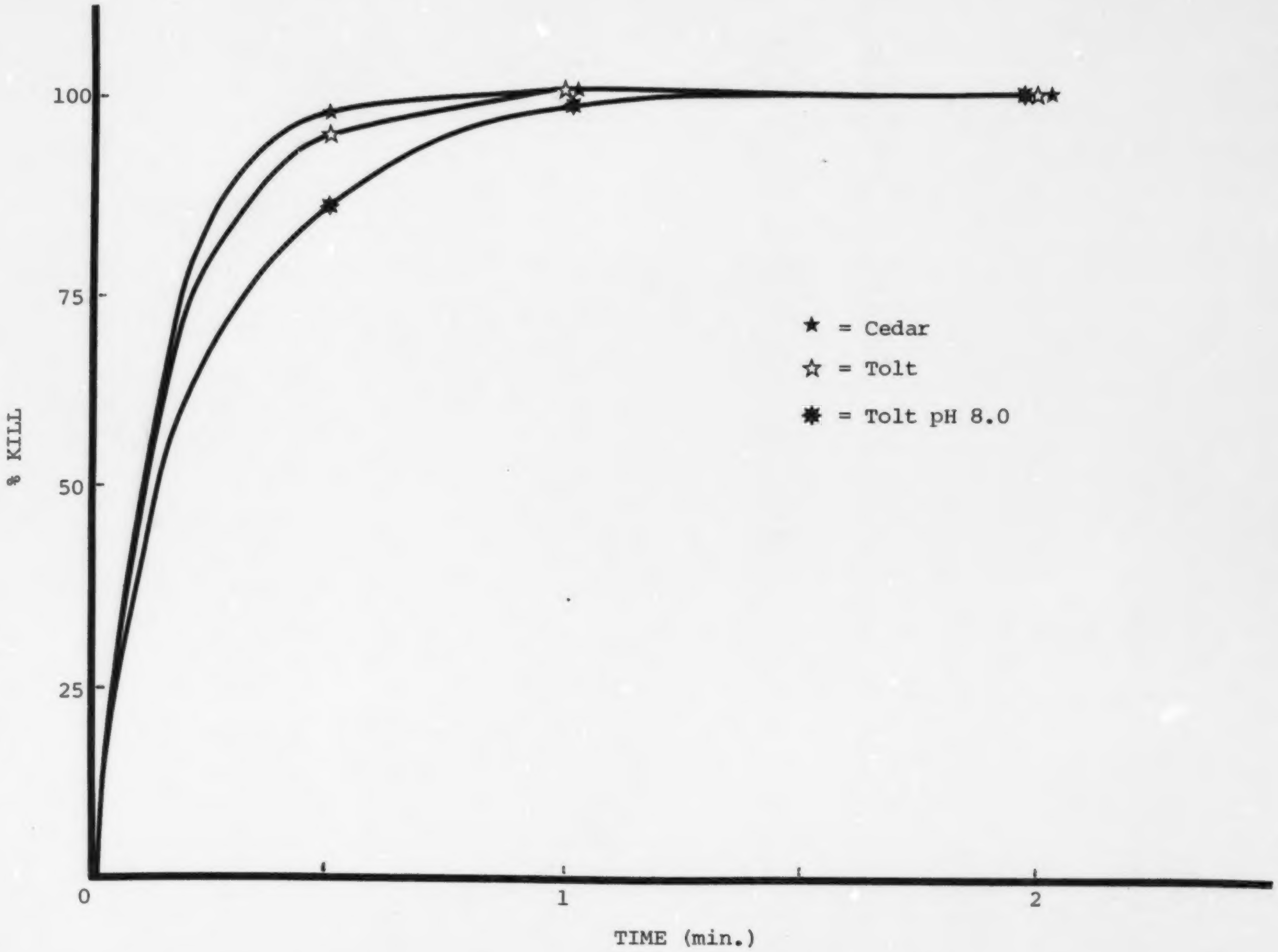

Figure 31. Percent kill of Enterobacter aerogenes at $[Cl_2]$ = 0.1 mg/L after 10 minutes.

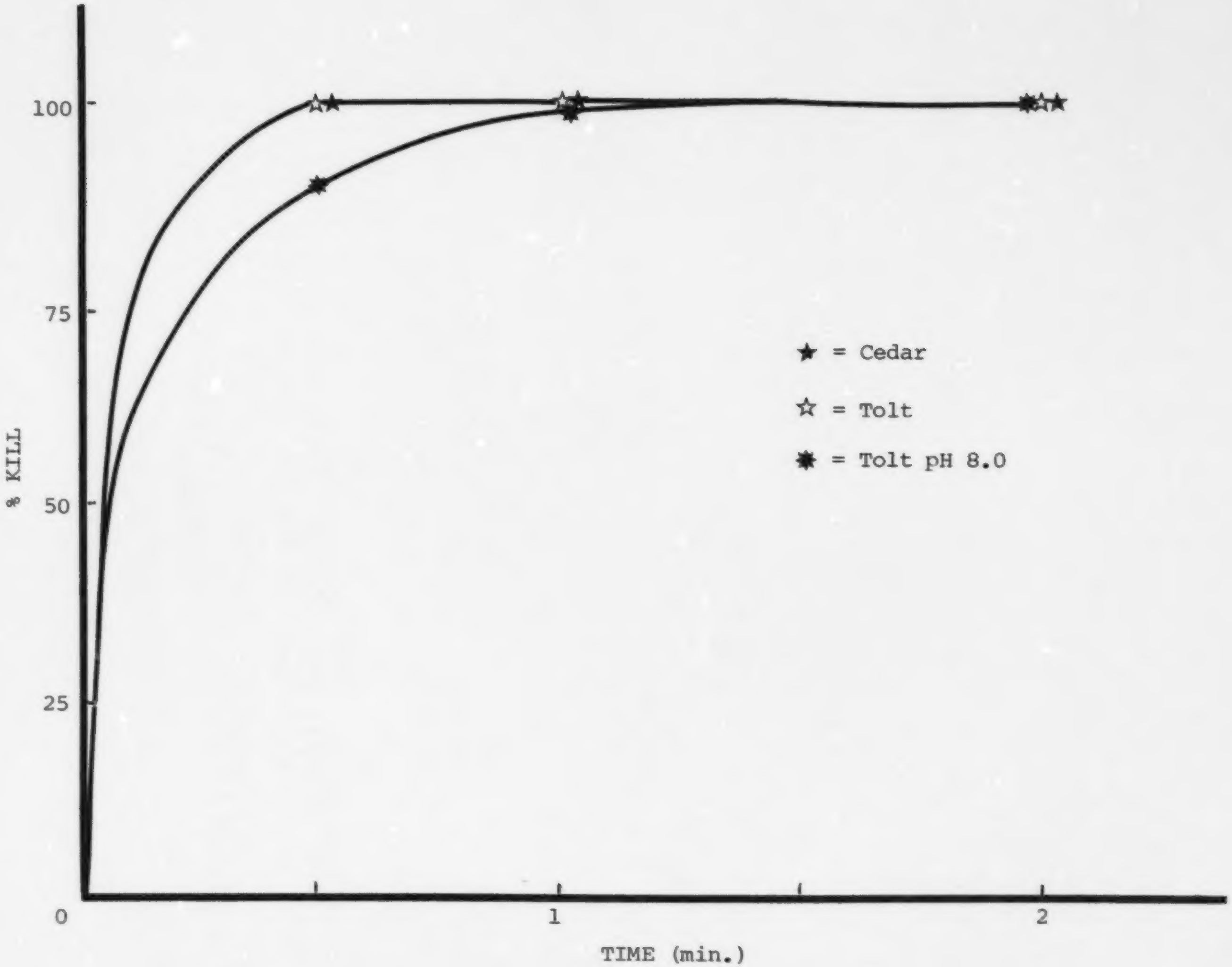

Figure 29. Percent kill of Enterobacter aerogenes at $[Cl_2]$ = 0.2 mg/L after 10 minutes.

TABLE 7. BACTERIAL CONCENTRATIONS OVER TIME FOR *PSEUDOMONAS AERUGINOSA*

Source	pH	Free Chlorine Residual mg/L	Contact Time, Minutes	Bacterial Concentration #/mL	Log Concentration
Tolt	6.47	0.0	-	4.50×10^4	4.67
	6.45	0.1 (dosage) 0.01 (residual)	0	2.50×10^4	4.39
			½	5.30×10^3	3.72
			1	5.20×10^3	3.70
			2	5.20×10^3	3.70
	6.47	0.12	0	4.50×10^4	4.65
			½	33	1.52
			1	33	1.52
			2	0	-
	6.42	0.28	0	4.38×10^4	4.64
			½	0	-
			1	0	-
			2	0	-
Cedar	7.09	0.0	-	4.69×10^4	4.67
	7.04	0.1 (dosage) 0.01 (residual)	0	4.69×10^4	4.67
			½	0	-
			1	0	-
			2	0	-
	7.05	0.24	0	4.69×10^4	4.67
			½	33	1.52
			1	33	1.52
			2	0	0
	7.11	0.34	0	4.71×10^4	4.67
			½	0	-
			1	0	-
			2	0	-
Tolt	8.03	0.0	-	4.61×10^4	4.66
	8.03	0.1 (dosage) 0.01 (residual)	0	2.80×10^4	4.45
			½	9.40×10^3	3.97
			1	9.20×10^3	3.96
			2	9.00×10^3	3.92
	8.03	0.15	0	4.61×10^4	4.66
			½	66	1.82
			1	33	1.52
			2	0	0
	8.03	0.25	0	4.62×10^4	4.66
			½	0	-
			1	0	-
			2	0	-

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TABLE 8. BACTERIAL CONCENTRATIONS OVER TIME FOR *ESCHERICHIA COLI*

Source	pH	Free Chlorine Residual mg/L	Contact Time, Minutes	Bacterial Concentration #/mL	Log Concentration
Tolt	6.48	0.00	-	7.6×10^4	4.88
	6.48	0.1 (dosage)	0	7.6×10^4	4.88
		0.02 (residual)	½	9.3×10^3	3.97
			1	8.4×10^3	3.92
			2	8.4×10^3	3.92
	6.48	0.23	0	7.6×10^4	4.88
			½	3.3×10^2	2.52
			1	0	-
			2	0	-
	6.48	0.37	0	7.6×10^4	4.88
			½	0	-
			1	0	-
			2	0	-
Cedar	7.29	0.00	-	7.20×10^4	4.86
	7.32	0.1 (dosage)	0	7.20×10^4	4.86
		0.00 (residual)	½	0	-
			1	0	-
			2	0	-
	7.23	0.10	0	7.20×10^4	4.86
			½	0	-
			1	0	-
			2	0	-
	7.24	0.15	0	7.10×10^4	4.85
			½	0	-
			1	0	-
			2	0	-
Tolt	8.01	0.00	-	7.10×10^4	4.85
		0.1 (dosage)	0	7.3×10^4	4.86
		0.01 (residual)	½	1.1×10^4	4.04
			1	1.2×10^4	4.07
			2	1.1×10^4	4.04
	8.14	0.17	0	7.00×10^4	4.84
			½	0	-
			1	0	-
			2	0	-
	8.00	0.21	0	7.1×10^4	4.85
			½	0	-
			1	0	-
			2	0	-

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TABLE 9. BACTERIAL CONCENTRATIONS OVER TIME FOR ENTEROBACTER AEROGENES

Source	pH	Free Chlorine Residual mg/L	Contact Time, Minutes	Bacterial Concentration #/mL	Log Concentration
Tolt	6.64	0.0	-	2.6×10^4	4.41
	6.50	0.1 (dosage)	0	2.7×10^4	4.43
		0.00 (residual)	½	4.0×10^3	3.60
			1	1.0×10^3	3.00
			2	0	0
	6.52	0.11	0	2.8×10^4	4.48
			½	1.5×10^3	3.17
			1	0	-
			2	0	-
	6.58	0.21	0	2.5×10^4	4.40
			½	0	-
			1	0	-
			2	0	-
Cedar	7.23	0.0	-	1.20×10^4	4.07
	7.20	0.1 (dosage)	0	1.8×10^4	4.26
		0.02 (residual)	½	6.6×10^2	2.82
			1	0	-
			2	0	-
	7.19	0.13	0	1.9×10^4	4.28
			½	6.6×10^2	2.82
			1	0	-
			2	0	-
	7.20	0.22	0	1.8×10^4	4.26
			½	0	-
			1	0	-
			2	0	-
Tolt	7.99	0.0	-	1.0×10^4	4.00
	8.17	0.1 (dosage)	0	1.0×10^4	4.00
		0.00 (residual)	½	2.7×10^3	3.43
			1	1.2×10^3	3.08
			2	1.0×10^3	3.00
	8.14	0.13	0	1.0×10^4	4.00
			½	1.4×10^3	3.14
			1	1.6×10^2	2.20
			2	0	-
	8.20	0.19	0	1.0×10^4	4.00
			½	1.1×10^3	3.04
			1	1.0×10^2	2.00
			2	0	-

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Effect of Contact Time on Disinfection

Figures 21 through 29 show the change in bacterial concentration and kill rates over time at the different chlorine concentrations and pH values. In other disinfection experiments in which bacteria were inoculated into sterile water containing a specific chlorine residual, the time to 100% kill was determined as a standard where percentage survival was plotted against time (10). In this study, however, the tests were designed to simulate natural conditions which occur in SWD's open reservoir system, through the use of natural waters and actual chlorination practices.

Hence, it was found that the time to 100% kill was somewhat variable as expected for tests subject to these conditions.[26] From Table 5 the time required for 100% kill ranged from one-half to one minute for pH 6; from one-half to one minute for pH 7; and from one to two minutes for pH 8. As expected, the time to 100% kill was greater at higher pH and less at higher concentrations of chlorine.

It should be noted that at pH 8 and a free chlorine residual of 0.2 mg/L the time to 100% kill was less than 30 seconds for P. aeruginosa and E. coli, and less than 2 minutes for E. aerogenes. These results point out that SWD's chlorination program, which maintains a free chlorine residual of 0.6 mg/L and a minimum 10-minute contact time, will be sufficient to provide adequate bacteriological disinfection.

ONSITE TREATMENT TESTS

General

The impact of treatment start-up on the microbial, chemical, and physical aspects of pipe loop water quality was determined as described in Section 5. The target and actual pH of each treatment start-up phase is presented in Table 10. These tests were conducted on old galvanized pipe because negative impacts due to treatment start-up are more likely to occur in galvanized pipe systems than in copper pipe systems. Previous studies conducted by SWD have shown little or no improvements in iron corrosion and greater metals leaching in some situations as a result of treatment.[2, 3] Also, the effects of corrosion on galvanized pipe such as colored water, turbidity, stained fixtures, and pipe deterioration, are much more apparent to the customer than the effects of copper corrosion.

TABLE 10. TREATMENT PHASES

Phase	Target pH	Actual Average pH	Duration
I	6.00	6.00	106 days
II	7.00	7.11	Day 0 to Day 37
III	7.60	7.37	Day 38 to Day 56
IV	8.00	8.08	Day 57 to Day 84

The Effects of Treatment Start-Up on Microbial Water Quality

Bacterial Plate Count --

It is impractical to attempt to enumerate all microorganisms that might be present in water because no single medium, temperature, or choice of incubation time can ensure recovery of all organisms present in water. However, certain members of the bacterial population in water can be measured, and their numbers can be related to the microbiological quality of the distribution system.

In an effort to develop a better cultivation method for enumerating microbial populations in the pipe loop samples, both incubation temperature and time were varied from the standard plate count protocol. In addition, if time had permitted, it would also have been informative to test a number of different media to determine their efficiency in supporting the various microbial populations in the premise plumbing systems.

Standard bacterial plate counts were incubated at 20-24°C for six days and compared to bacterial plate counts incubated for 48 hours at 35°C (Figure 30). The extended incubation time showed an average increase of as much as two orders of magnitude of microorganisms per mL of sample over the shorter incubation period.

It is evident that the lower incubation temperatures (20-24°C) are more closely related to the actual water temperatures to which the microorganisms are accustomed. Alternatively, it might be that the lower temperatures may be a prerequisite to better viability of indigenous microbial populations on artificially enriched laboratory media, such as plate count medium.

Major Biochemical Groups Isolated in Study --

A total of 2210 colonies were picked from standard plate count agar incubated at 20-24°C and 35°C. The isolates were assigned to arbitrary numbered groups on the basis of the pattern obtained with the seven tests described in Section 5. A total of 24 groups were found within the 2210 isolates. The 17 major groups, comprising 90% of the total isolates tested, are shown in Table 11. The groups are ranked in decreasing order of predominance at 35°C. Gram reaction, nitrate reduction, citrate utilization, and growth on MacConkey agar were determined for a portion of the isolates in each gorup to obtain further information and to evaluate homogeneity within the group. A further description of the groups follows.

Gram Negative Aerobic Nonsaccharolytic Rods --

Group 5--Gram stains were performed on 54 isolates in Group 5, of which 52 (96%) were Gram negative rods, 2 (3%) were Gram positive rods, and 1 (1%) were Gram positive cocci. Yellow pigmentation was common within Group 5, with 45% of the isolates at 20-24°C and 58% of the isolates at 35°C having this characteristic. The results for the additional tests for Group 5 are shown in Table 12.

Group 5 appears to be fairly homogenous, with 61% of the members of this group showing one pattern on the three tests.

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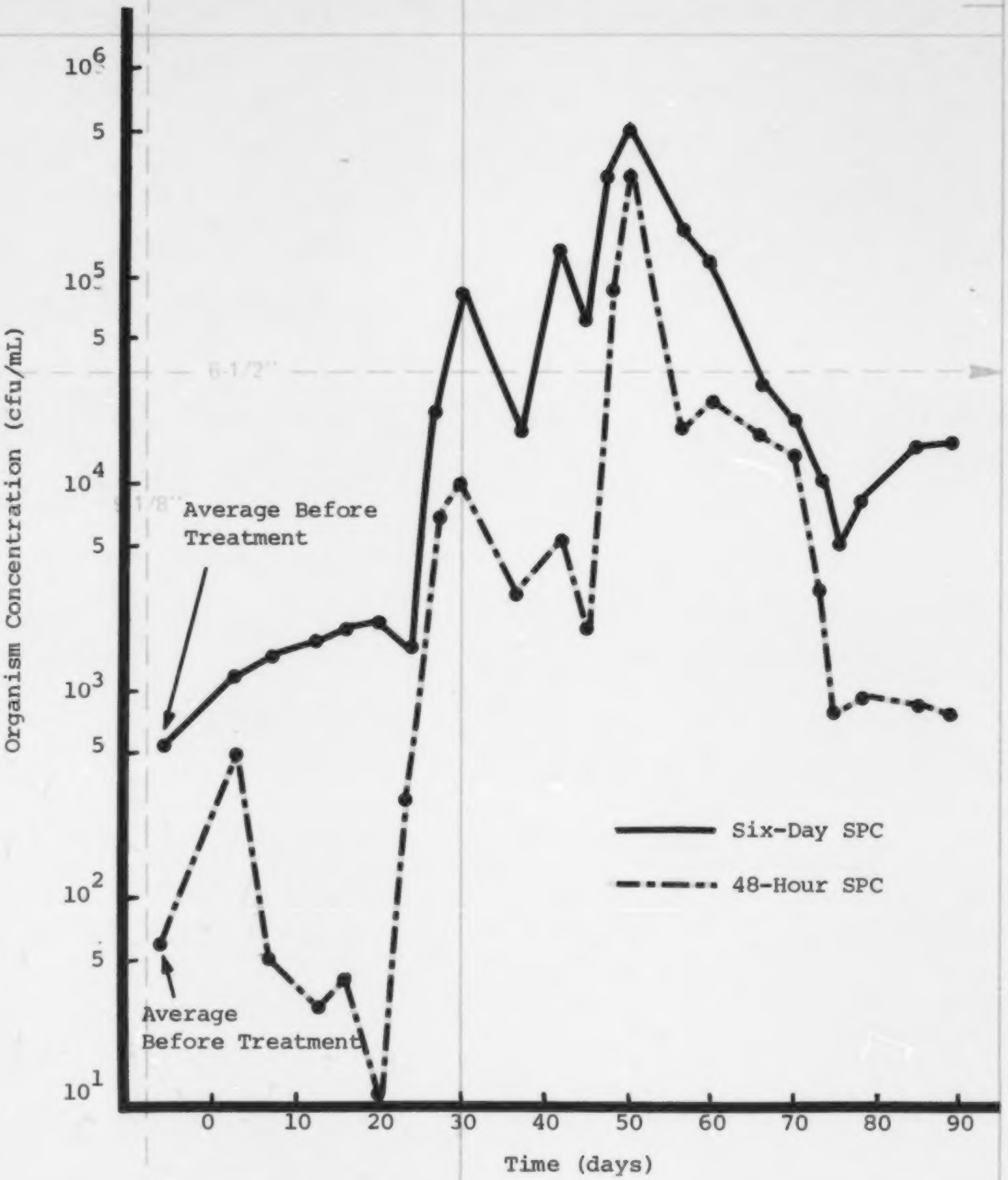

Figure 30. Comparison of 48-hour and six-day standard plate count for standing water.

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TABLE 11. MAJOR BIOCHEMICAL GROUPS ISOLATED FROM 24-HOUR STANDING WATER SIX DAY BACTERIAL PLATE COUNTS FROM PILOT PLANT GALVANIZED PIPE LOOP SYSTEM.

Group	Facultative Anaerobe	Catalase Activity	Oxidase	Motility	Indole	Acid from Glucose Anaerobically	Acid from Glucose Aerobically	20-24°C Number Isolated (% of Total)		35°C Number Isolated (% of Total)	
5	-	+	+	-	-	-	-	253	(23.0)	164	(18.5)
18	-	+	-	-	-	-	-	348	(31.6)	133	(15.0)
4	-	+	+	+	-	-	-	53	(4.8)	90	(10.1)
10	+	-	-	-	-	-	-	79	(7.2)	87	(9.8)
8	+	+	+	+	+	+	+	11	(1.0)	85	(9.6)
13	+	+	-	+	-	-	-	79	(7.2)	78	(8.8)
9	+	+	-	-	-	-	-	59	(5.3)	48	(5.4)
23	-	+	+	-	-	-	+	21	(1.9)	36	(4.1)
19	-	+	-	+	-	-	-	42	(3.8)	24	(2.7)
15	+	+	+	+	-	-	-	31	(2.8)	23	(2.6)
7	+	-	-	-	-	+	-	11	(1.0)	19	(2.1)
1	+	+	+	+	-	+	+	25	(2.3)	13	(1.5)
6	-	+	-	-	-	-	+	15	(1.4)	12	(1.3)
11	+	+	-	+	-	+	+	3	(.3)	10	(1.1)
20	+	+	-	-	-	+	+	2	(.2)	9	(1.0)
2	-	+	+	+	-	-	+	9	(.8)	7	(.8)
3	-	+	-	+	-	-	+	10	(.9)	4	(.5)
							TOTAL	1102	(95.5)	887	(93.9)

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TABLE 12. GRAM NEGATIVE AEROBIC NONSACCHAROLYTIC RODS

Group	Nitrate Reduction	Growth on MacConkey	Citrate Utilization	Number Obtained / Number Tested	%
5	-	-	-	17/28	61
	+	-	-	6/28	21
	+	+	+	3/28	11
	-	+	+	2/28	7
18	-	-	-	24/38	63
	+	+	+	6/38	16
	+	+	-	4/38	11
	+	-	-	2/38	5
	-	+	+	1/38	3
	-	-	+	1/38	3
4	-	-	-	11/15	73
	+	+	-	3/15	20
	+	-	-	1/15	7
19	-	-	-	7/10	70
	-	+	+	2/10	20
	+	+	+	1/10	10

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Group 18--Group 18 was composed predominantly of Gram negative rods with 64 (89%) of the isolates examined falling into this category while 4 (6%) were Gram positive cocci, and 4 (5%) were Gram positive rods. On the basis of three additional tests, this group was also fairly homogeneous, as shown in Table 12.

64% of the isolates at 20-24°C were yellow, while 42% at 35°C were yellow.

Group 4--Of the 24 isolates in Group 4 examined for Gram stain, 21 (89%) were Gram negative rods and 3 (11%) were Gram positive rods. Yellow pigmentation was less prevalent in this group, with 14% of the isolates at 20-24°C and 12% at 35°C showing this pigmentation. Three patterns were obtained with the three additional tests, as shown in Table 12. The triple negative pattern was again the most prevalent, with 73% of the isolates examined in Group 4, as shown in Table 12.

Group 19--Since this group composed a smaller fraction of the total number of isolates, fewer members of this group were examined. 20 isolates were Gram stained, of which 13 (65%) were Gram negative rods, 4 (20%) were Gram positive rods, and 3 (15%) were Gram positive cocci. Considerable heterogeneity was found for nitrate reduction, growth on MacConkey agar and utilization of citrate. 48% of the isolates at 20-24°C, and 62% of the isolates at 35°C demonstrated yellow pigmentation.

Although positive identification of bacteria in these groups cannot be made on the basis of the limited number of tests utilized, a tentative identification is possible. Possibilities for groups 5, 18, 4, and 19 include Pseudomonadaceae, Flavobacterium, Acinetobacter, Alcaligenes, and Moraxella.

Gram Negative Facultative Nonsaccharolytic Rods --

Group 10--Group 10 was composed of 97% (35/36) Gram negative rods and 3% Gram positive rods. Approximately half of the isolates in this group were pigmented yellow, 48% at 20-24°C and 53% at 35°C. Results of the nitrate reduction, growth on MacConkey, and citrate utilization are shown in Table 13.

Group 13--This group was composed almost entirely of yellow pigmented Gram negative rods. Of the 28 isolates examined for Gram stain, 26 (93%) were Gram negative rods and 2 (1%) were Gram positive rods. 68% of the isolates at 20-24°C showed orange pigmentation and 56% at 35°C showed yellow pigmentation. This group was homogeneous with respect to nitrate reduction, growth on MacConkey and citrate utilization with the triple negative pattern predominant.

Group 9--Group 9 was similar to Group 13 in that many of the isolates showed orange pigmentation, 72% at 20-24°C, and yellow pigmentation 58% at 35°C. 79% of isolates (19/24) were Gram negative rods, 17% (4/24) were Gram positive cocci, and 4% (1/24) were Gram positive rods. The results for nitrate reduction, growth on MacConkey, and citrate utilization indicate that this group is homogeneous with respect to these tests.

TABLE 13. GRAM NEGATIVE FACULTATIVE NONSACCHAROLYTIC RODS

Group	Nitrate Reduction	Growth on MacConkey	Citrate Utilization	Number Obtained / Number Tested	%
10	-	-	-	8/12	67
	+	-	-	4/12	33
13	-	-	-	18/24	75
	+	+	+	3/24	13
	+	-	-	1/24	4
	-	+	-	1/24	4
	+	+	-	1/24	4
9	-	-	-	11/14	79
	-	-	+	1/14	7
	+	-	-	1/14	7
	+	+	+	1/14	7
15	-	-	-	7/11	64
	+	-	-	3/11	27
	-	+	-	1/11	9

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TABLE 14. SACCHAROLYTIC MICROORGANISMS

GRAM NEGATIVE FACULTATIVE SACCHAROLYTIC RODS

Group	Nitrate Reduction	Growth on MacConkey	Citrate Utilization	Number Obtained / Number Tested	%
8	+	+	+	6/6	100
1	+	+	+	3/3	100

GRAM POSITIVE OR NEGATIVE AEROBIC SACCHAROLYTIC RODS

Group	Gram Reaction		% Yellow Colonies	
	Positive Rod	Negative Rod	20-24°C	35°C
23	4	9	15	7
6	7	4	24	9
2	26	4	0	0
3	14	6	4	0

FACULTATIVE SACCHAROLYTIC MICROORGANISMS

Group	Gram Positive Rod	Gram Negative Rod	Gram Positive Coccus	Gram Positive Filament
7	4	3	0	12
11	7	0	15	0
20	0	9	0	0

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Group 15--Group 15 contained 95% (17) Gram negative rods and 5% (11) Gram positive rods. This group differed from the other three in this category in that it contained a low percentage of yellow pigmented colonies, 38% at 20-24°C and 21% at 35°C, and showed greater diversity in the three additional tests run. The majority of bacteria in groups 10, 13, 9, and 15 were consistent with the biochemical characteristics of *Flavobacterium*.

Gram Negative Facultative Saccharolytic Rods --

Groups 8 and 1--Isolates in these two groups were characterized by positive reactions on most of the biochemical test runs. A total of 23 isolates were examined for Gram reaction and morphology; 15 in Group 8 and 8 in Group 1. All but 2 were Gram negative rods. Yellow pigmentation was generally absent; 2% at 20°C and 6% at 35°C for Group 8, and 4% at 20-24°C and 0% at 35°C for Group 1.

Gram Positive or Negative Aerobic Saccharolytic Rods --

Groups 23, 6, 2, and 3--The aerobic-saccharolytic category contained both Gram positive and Gram negative rods and a low percentage of pigmented colonies as shown in Table 14.

In addition, 4 Gram positive cocci were obtained in Group 23 and 10 Gram positive cocci were obtained in Group 6. Since the 4 groups accounted for only 5.2% of the total number of isolates, they were not tested for nitrate reduction, growth on MacConkey, and citrate utilization. The Gram positive organisms in Group 23 showed characteristics consistent with *Bacillus*, while those in Group 6 are most probably *Micrococcus*. The Gram negative organisms can be tentatively identified as *Pseudomonadaceae*.

Facultative Saccharolytic Microorganisms --

Groups 7, 11, and 20--These groups differed from the preceding groups in that they contained a higher percentage of Gram positive microorganisms. Yellow pigmentation in the groups varied. Tentative identification for the Gram positive filaments may be *Actinomyces*, the Gram positive rods as *Bacillus*, the Gram positive cocci as *Staphylococcus*, while the Gram negative rods may be *Enterobacteriaceae*.

Summary of Biochemical Tests --

The 17 major groups shown in Table 15 are ranked in the order of predominance at 35°C. The trend obtained at 20-24°C was generally the same as is also shown in Table 15. Table 15 lists the SPC bacteria tentatively isolated from the pipe loop system. Groups 5 and 18 were isolated in the greatest number, with 54.6% of the total isolates at 20-24°C and 33.5% of the total isolates at 35°C belonging to these two groups.

The majority of isolates collected in this study were Gram negative nonsaccharolytic rod-shaped bacteria. The majority of microorganisms in this category were obligate aerobes but significant numbers of facultative bacteria were also observed. Both the aerobic and facultative groups contained a large percentage of microorganisms that yielded pigmented colonies. The biochemical characteristics of the aerobic group are consistent with the members of the family *Pseudomonadaceae* and the genera *Flavobacterium*, *Alcaligenes*,

TABLE 15. SUMMARY OF SPC BACTERIA TENTATIVELY ISOLATED FROM 24-HOUR STANDING PIPE LOOP WATER.

Organism	Group	Category
Pseudomonas, *Flavobacterium*, *Alcaligenes*, *Acinetobacter*, *Moraxella*	5, 18, 4, 19	Gram negative aerobic nonsaccharolytic rods.
Flavobacterium	10, 13, 9, 15	Gram negative facultative nonsaccharolytic rods.
Aeromonas	8, 1	Gram negative facultative saccharolytic rods.
Pseudomonas, *Bacillus*, *Micrococcus*	23, 6, 2, 3	Gram positive or negative aerobic saccharolytic rods.
Actinomyces, *Staphylococcus*, *Bacillus*, *Enterobacter*	7, 11, 20	Facultative saccharolytic microorganisms.

and *Moraxella*, and those of the facultative groups are consistent with members of the genus *Flavobacterium*. Phases II and III (Figures 35 and 36) displayed the greatest variety in types of microorganisms present. Scanning electron micrographs of various SPC bacteria are shown in Appendix C.

Group 8 (possibly *Aeromonas*) and Groups 11 and 20 (possibly *Enterobacteriaceae*, *Bacillus* and/or *Staphylococci*) were found almost exclusively at 35°C during Phases II and III (Figures 32 and 33).

Groups 5, 18, and 19 (possibly *Pseudomonadaceae* and *Flavobacterium* predominating) showed a significant increase in growth at 20-24°C as opposed to 35°C. In addition, these organisms increased in numbers with corrosion treatment (see Figures 31 through 34).

Bacteria that may be primary or opportunistic pathogens (*Pseudomonas aeruginosa* and other *Pseudomonas spp*., as well as *Flavobacterium spp*., *Acinetobacter spp*., *Moraxella spp*., *Staphyloccocus aureus* and *Aeromonas*)[19, 31, 33] comprised approximately 77% of the total SPC population identified. Populations of *Pseudomonas aeruginosa* and *Flavobacterium* have been recognized as health hazards to patients in burn centers, nurseries, hospitals, and nursing homes. *Aeromonas spp*. have also been associated with a wide range of human infections, indicating that these organisms may act as primary pathogens.[33] In addition, many of these bacteria, including *Pseudomonas*, *Flavobacterium*, *Moraxella*, and *Acinetobacter*, have been found to be multiply antibiotic resistant (Appendix E). It is believed that these drug-resistant organisms are becoming more common due to the extensive use of antibiotics in medicine and agriculture throughout the world.

Bacteria considered antagonistic to coliforms (including various species of *Pseudomonas*, *Flavobacterium*, *Bacillus*, *Micrococcus*, and *Acinetobacter*[32, 34]) comprised approximately 71% of the total SPC population identified. In addition, a group of filamentous bacteria called *Actinomycetes*, also considered to be coliform antagonists, was tentatively identified in large numbers, both in the standing water and on surfaces of pipe incrustations. No definitive quantitative studies were performed concerning these organisms.

Geldreich, *et*. *al*.[35] noted the significance of coliform suppression in water supplies and observed that suppression can at least in part be attributed to elevated SPC numbers. For example, increased SPC densities have been associated with waterborne outbreaks.[35, 36] Pathogen occurrences have also been observed in the absence of detectable coliforms in samples with high SPC numbers.[32]

Microbial Quality of Pipe Loop Water During Treatment Start-Up --

Mean standard plate count levels, turbidity, iron and zinc levels were all significantly elevated in 24-hour standing water samples (Table 16). Each phase showed a difference of two to three orders of magnitude of microorganisms per mL between standing and running samples for SPC bacteria.

As indicated previously in Figure 30, the extended incubation time of six days at 20-24°C showed an increase of as much as two orders of magnitude of microorganisms per mL of sample over the shorter incubation time. Figure 35

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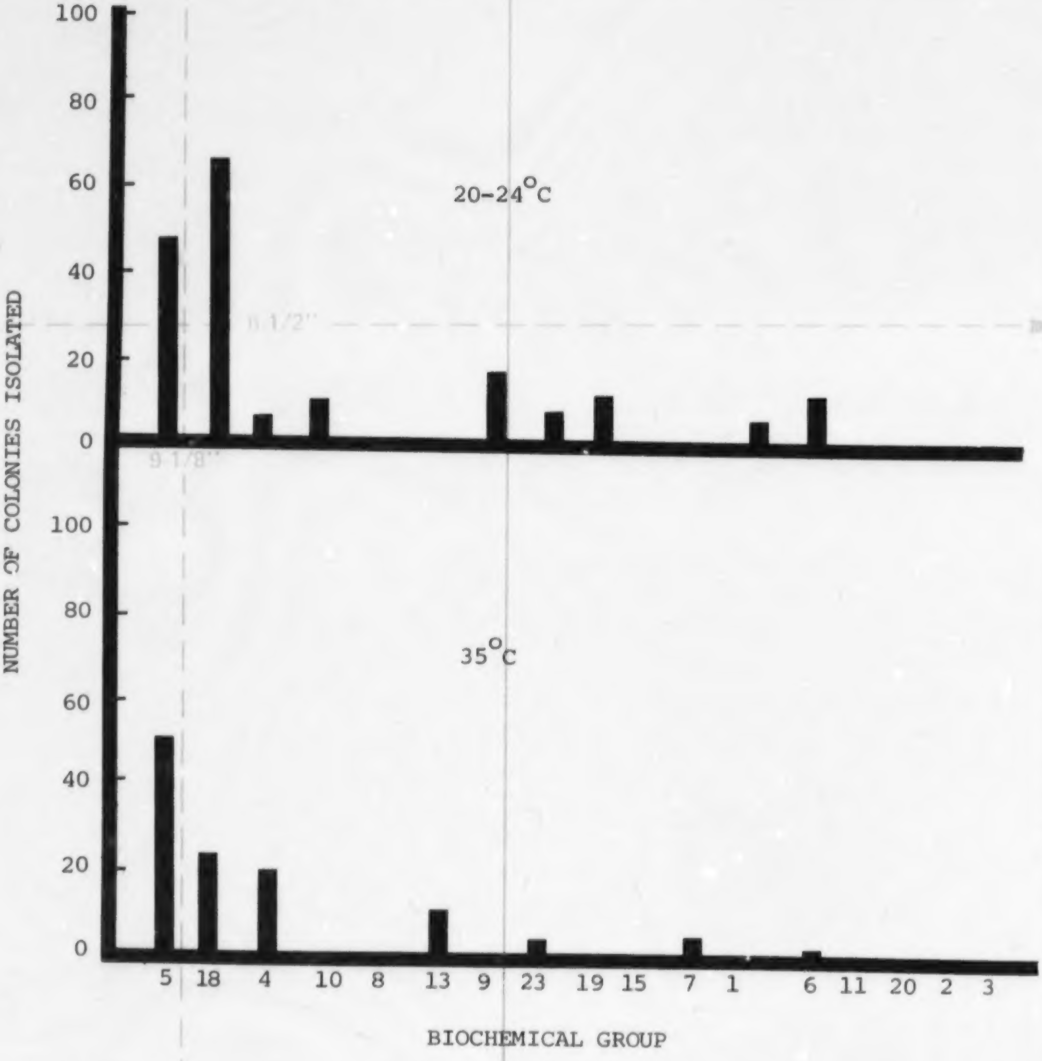

Figure 31. Phase I: Occurrence of the different biochemical groups in 24-hour standing water.

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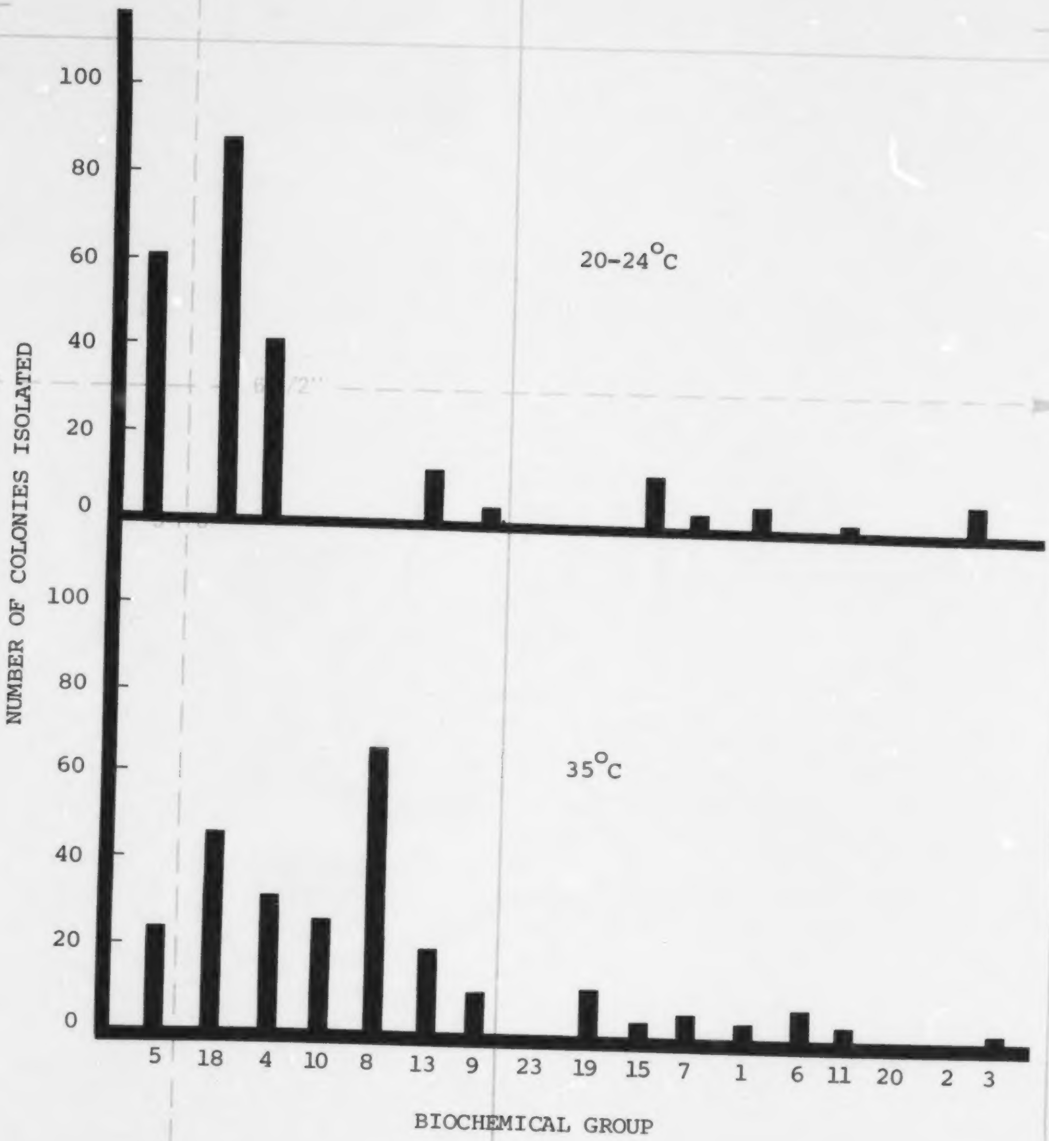

Figure 32. Phase II: Occurrence of the different biochemical groups in 24-hour standing water.

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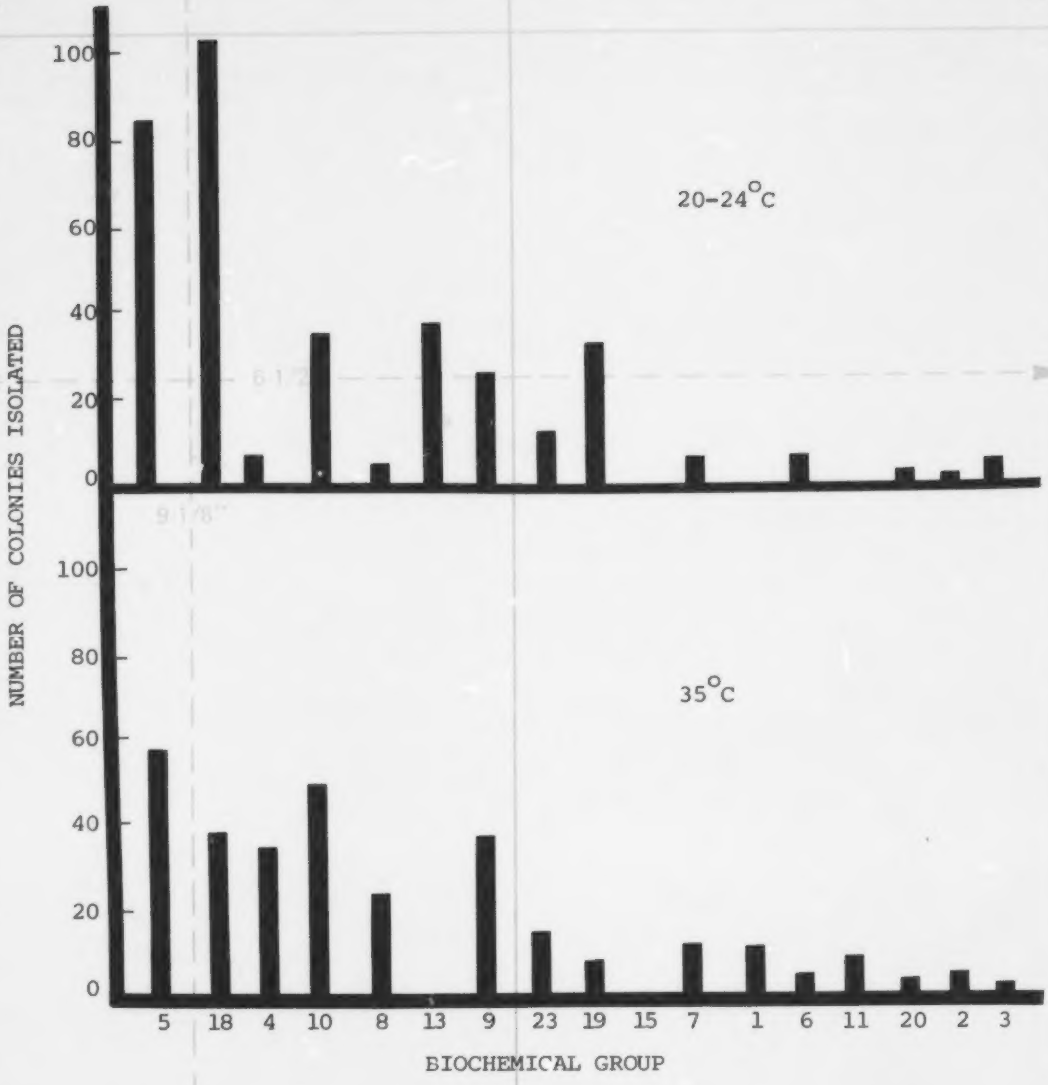

Figure 33. Phase III: Occurrence of the different biochemical groups in 24-hour standing water.

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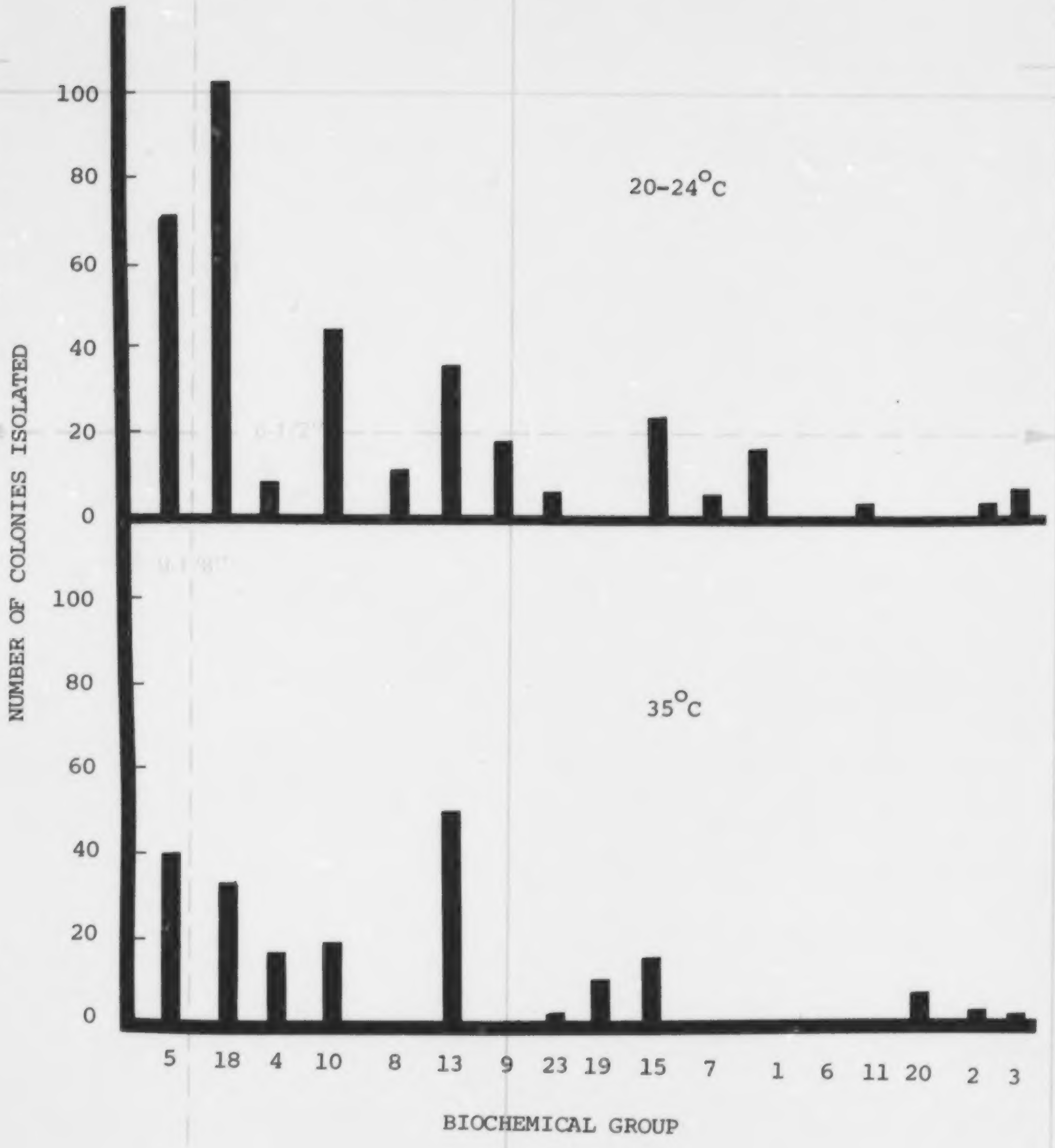

Figure 34. Phase IV: Occurrence of the different biochemical groups in 24-hour standing water.

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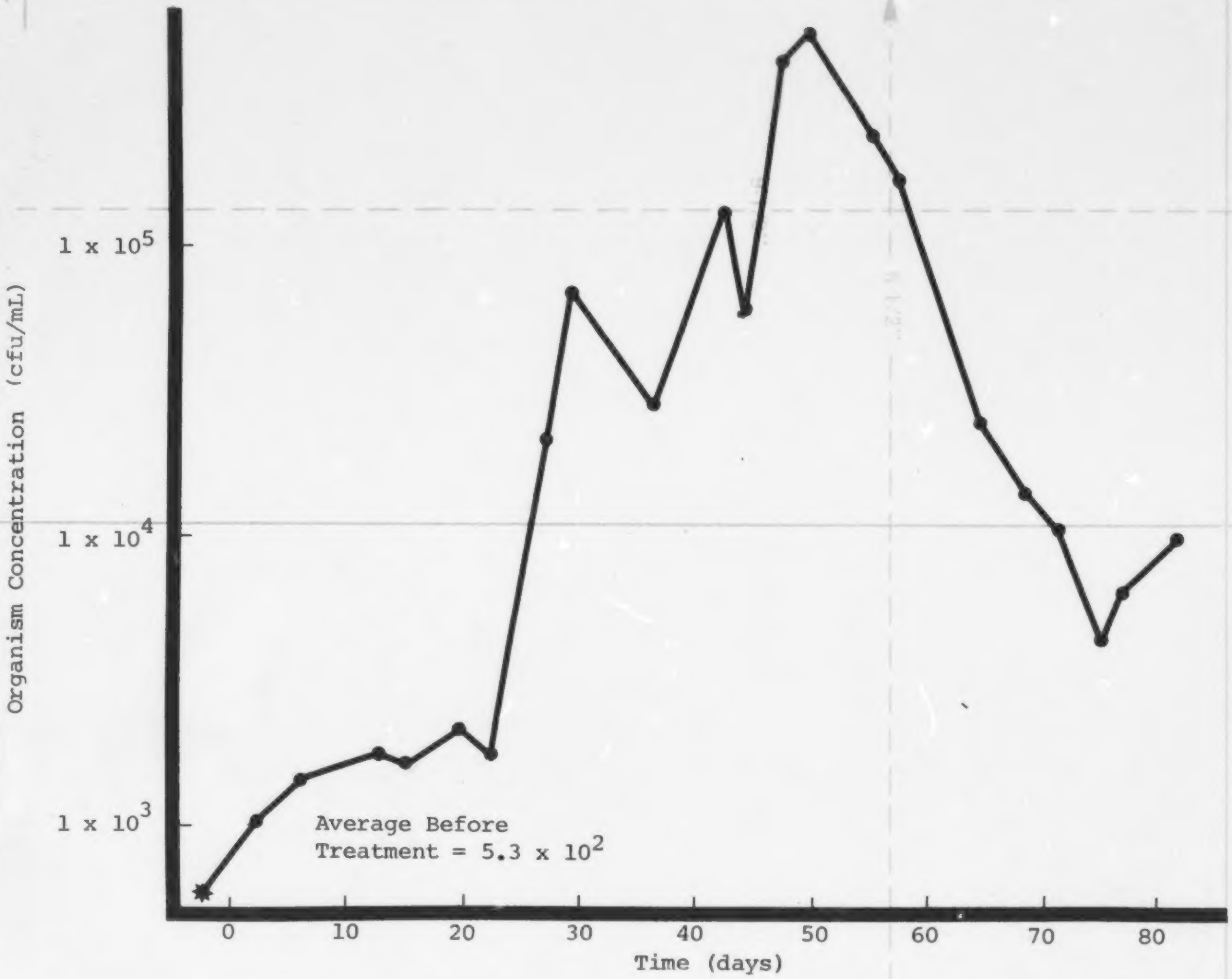

Figure 35. Standing water six-day standard plate count versus time.

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TABLE 16. MEANS OF SELECTED PARAMETERS BEFORE AND AFTER CORROSION TREATMENT START-UP.

Phase	Standard Plate Count (cfu/mL) 48 hours @ 35°C	Standard Plate Count (cfu/mL) 6 days @ 20-24°C	Coliforms (cfu/100 mL)
Phase I (13 samples)			
Standing	60.0	620	<0.5
Running	9.0	86.0	<0.5
Control	1.0	3.0	<0.5
Phase II (9 samples)			
Standing	2.23×10^3	1.51×10^4	9.0
Running	46.0	1.38×10^2	<0.5
Control	<0.5	2.0	<0.5
Phase III (4 samples)			
Standing	1.48×10^5	2.55×10^5	1.0
Running	1.35×10^2	8.80×10^2	0.5
Control	1.0	7.0	0.5
Phase IV (9 samples)			
Standing	1.33×10^4	4.32×10^4	6.0
Running	1.20×10^2	4.13×10^2	1.0
Control	8.0	14.0	<0.5

(continued)

TABLE 16, Cont.

Phase	Turbidity (NTU)	Fe (mg/L)	Zn (mg/L)	Free Cl_2 (mg/L)
Phase I (13 samples)	Mean ± S.D.	Mean ± S.D.	Mean ± S.D.	
Standing	6.2 ± 3.1	3.95 ± 1.94	4.67 ± 0.99	0
Running	1.2 ± 0.15	0.420 ± 0.27	0.310 ± 0.10	0.20
Control	1.1 ± 0.17	0.213 ± 0.115	0.049 ± 0.036	0.20
Phase II (9 samples)				
Standing	8.0 ± 1.2	5.52 ± 0.46	1.82 ± 0.53	0
Running	0.58 ± 0.12	0.166 ± 0.06	0.13 ± 0.11	0.15
Control	0.51 ± 0.08	0.071 ± 0.016	0.020 ± 0.007	0.15
Phase III (4 samples)				
Standing	20.0 ± 5.6	6.67 ± 2.25	1.80 ± 0.87	0
Running	0.44 ± 0.01	0.140 ± 0.07	0.096 ± 0.068	0
Control	0.44 ± 0.10	0.060 ± 0.008	0.016 ± 0.007	0
Phase IV (9 samples)				
Standing	17.0 ± 2.4	6.00 ± 0.73	2.13 ± 0.56	0
Running	0.51 ± 0.11	0.320 ± 0.21	0.087 ± 0.039	0
Control	0.41 ± 0.21	0.080 ± 0.016	0.023 ± 0.009	0

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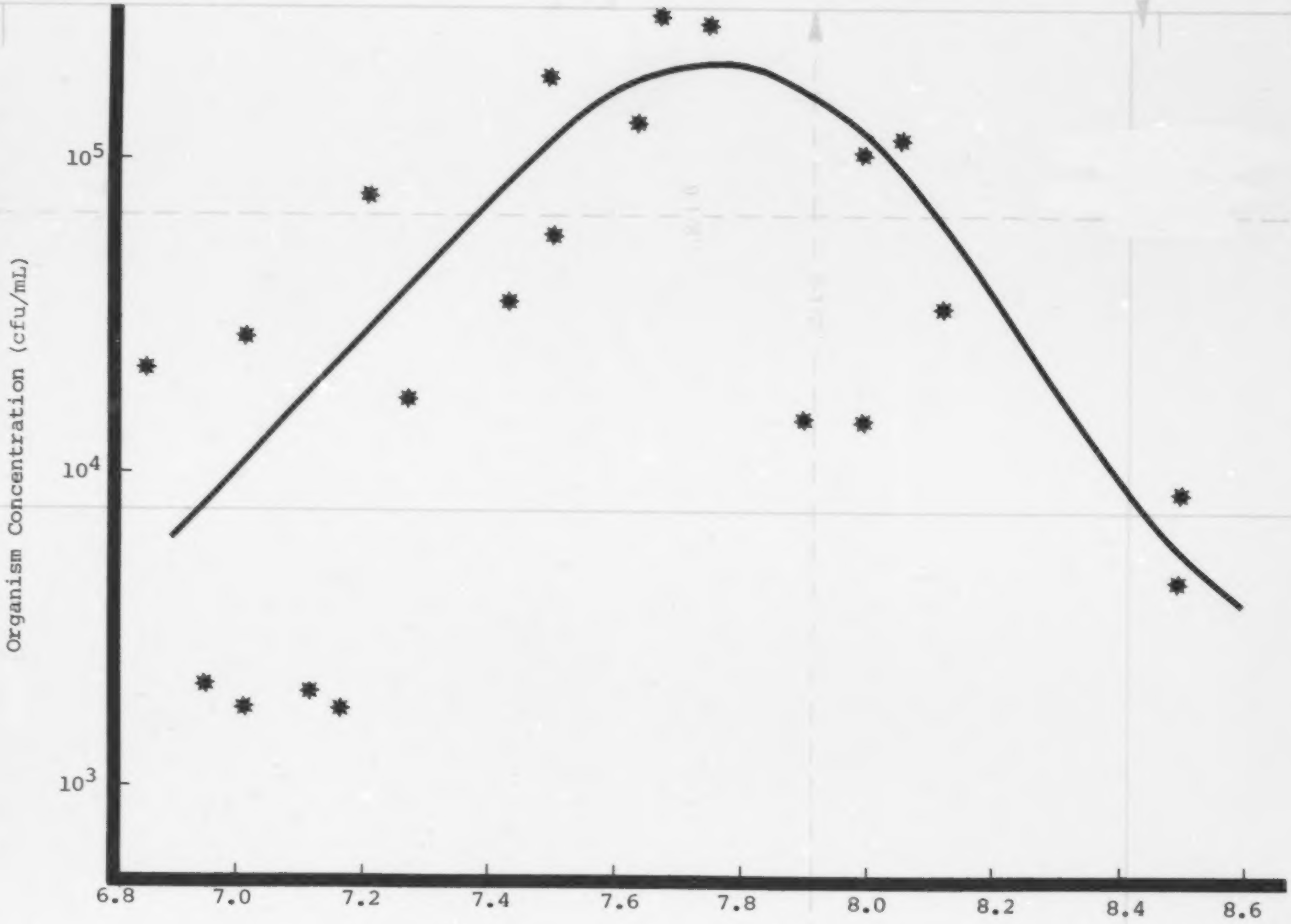

Figure 36. Standing water standard plate count versus pH.

is an enlarged version of the six-day standard plate count presented in Figure 30. The highest bacterial counts/mL resulted between day 30 and day 60, i.e., starting the end of Phase II and continuing through the beginning of Phase IV with an average of 10^5 org/mL in standing water samples.

Figures C-5 through C-22 show representative scanning electron micrographs of microorganisms and debris present in water samples collected from the pipe loop prior to and during each phase of corrosion treatment. Samples of 24-hour standing water always showed a greater accumulation of organic and inorganic material as well as greater numbers of microorganisms in contrast to the running and control water samples. This was especially apparent during the course of corrosion treatment.

A definite increase was observed in the number of heterotrophic bacteria which can precipitate iron from solution. The presence of aggregated bacteria encrusted with ferric hydroxide, instead of the usual filamentous iron bacteria, have been found in iron precipitates. Genera which contain species that have been found capable of precipitating iron from the salts of organic acids include Enterobacter, Bacillus, Pseudomonas, Serratia, Flavobacterium, Mycobacterium and Hyphomicrobium. The ferric hydroxide is apparently a waste product and iron salts are a necessary metobolite for these forms. Microorganisms may also produce a reducing environment by forming metabolite end products which can act as reductants of iron.[37, 38, 39]

A plausible explanation for these results may be that due to the prevailing chemical and physical conditions such as pH, turbidity, and trace metal availability, changes in the microbial population occurred. This allowed certain microorganisms to predominate, such as filamentous bacteria or organisms which shed exopolysaccharide fibers, etc., thus creating protected habitats which act as places of entrapment and harbor items such as corrosion products as well as provide a biological graveyard for cells of bacteria, algae, and diatoms. In these areas, bacteria are often afforded protection from disinfectant action, especially in premise plumbing systems where there are often long standing times and nonexistent chlorine residuals. In addition, the sediments which can accumulate in these areas provide the organic nutrients to sustain microbial life and encourage selective regrowth of certain bacteria in the microbiota. Some of these regrowth events often contribute organisms that suppress detection of low-density levels of total coliforms,[39] and other organisms capable of causing corrosion or taste and odor problems.

Figure 36 illustrates the effects of pH on six-day standard plate counts of 24-hour standing water. A steady increase in the number of microorganisms per mL was observed beginning at pH 6.8 and rising to approximately 7.0, then declining to pH 8.5. One explanation for this may be the fact that the optimum pH for most of the microorganisms of concern in this study is between approximately 6.5 and 7.8.[40]

Turbidity in 24-hour standing water after treatment start-up ranged from 6.2 NTU in Phase II to 25.0 NTU in Phase III. A mean of 8.0 NTU was received during Phase II, while Phases III and IV exhibited an average mean of 18.5 NTU which was significantly correlated with six-day standard plate count numbers of 24-hour standing water (Figure 40). This association may be due to a shielding effect, with collodial particles shielding microorganisms from the disinfectant.[41] However, since no chlorine residual was present during treatment start-up, as is sometimes the case in premise plumbing systems, this association may also be due to microbial action. Significant growth of iron precipitating bacteria can enhance iron flocculation due to excretion of extracellular polymers.[37] Thus turbidity may be either a cause or an effect of increased bacterial levels.

Large numbers of branched or forked microorganisms tentatively identified as Actinomycetes were frequently observed in the pipe loop water, most noticeable during Phases II and III (see Figures C-12 and C-23 through C-25).

Large numbers of rod-shaped and coccoid bacteria were viewed adhering to the surfaces of suspended particulate matter* and diatoms recovered from water samples via filtration with a substantial increase observed during Phases III and IV. In the majority of instances, the bacteria were clearly attached in a superficial fashion to the inside or outside of the particle and sometimes appeared to be covered by an extracellular slime layer (Figures C-26 to C-28). Biosynthesis of similar extracellular muccopolysaccharide substances by Pseudomonas aeruginosa has been shown to be an important factor in decreasing the susceptibility of this microorganism to chlorine disinfection (see Appendix C, for pathogenic effects of P. aeruginosa). In other instances, the cells were entrained within a mass of extracellular fibrillar or muccoid-appearing material (Figures C-29 and C-30).

Pseudomonas aeruginosa counts increased 57% from Phase I to Phase III (see Table 17), while other Pseudomonas spp., Pseudomonas-like bacteria, and Flavobacterium also increased in number with corrosion treatment (see Figures 31 through 34).

In addition to SPC bacteria, other microorganisms, some directly corrosion related, were observed in the pipe loop through the use of microbiological isolation techniques and scanning electron microscopy.

Many different species of diatoms and algae were observed with SEM as well as species of Cyanobacteria (see Figures C-26 and C-32 through C-34).

Stalk-producing organisms were observed frequently, especially tentatively identified species of Caulobacter (budding, stalked iron bacteria)

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*The precise chemical composition of these particles is not known. However, evidence from X-ray energy-dispersive microanalysis of suspended particulates suggests that a significant proportion ($>30\%$) may be organic and thus represents microscopic detritus particles introduced into the water system (Figures C-31 and C-55).

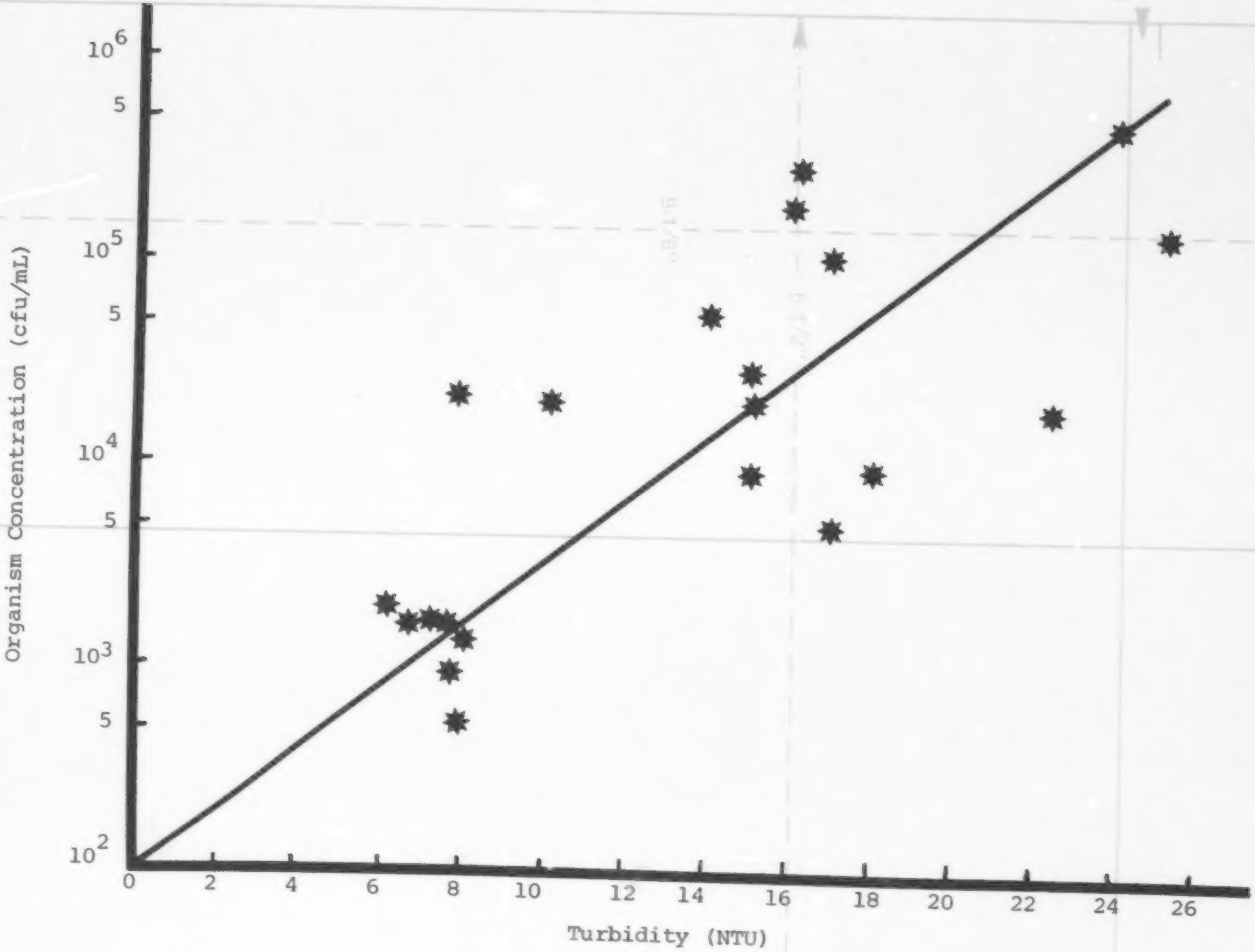

Figure 37. Standing water six-day standard plate counts versus turbidity.

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TABLE 17. PRESENCE OF PSEUDOMONAS AERUGINOSA IN PIPE LOOP WATER DURING TREATMENT START-UP

Phase	Pipe Loop Water (org/100 ml) Control	Standing	Tubercles
I	<0.5	32	Not detected.
II	1.0	64	Not examined.
III	1.0	74	Not examined.
IV	<0.5	44	Not detected.

and Hyphomicrobium (see Figures C-18, C-35 through C-38, and Table 18). These were often found entwined with and attached to algal cells, while others were attached to various bacteria or inorganic and organic debris (see Figure C-39). The survival efficiency of stalk-producing organisms lies in their ability to maintain viability under an adverse condition of low organic concentrations in water. This may be an important group of microorganisms since they can attach to themselves (forming "rosettes") or to other organisms and establish a biological network. Such attachment may enhance the formation of biological accumulation on the pipe surface. In addition, these sessile populations of microorganisms have been shown to be metabolically active and to contribute to the total microheterotrophic activity of certain aquatic ecosystems.[42]

Seliberia, and iron-transforming microorganism, distinguished by its rod-shaped, spirally twisted morphology, is tentatively identified as the organism in Figure C-17. Although a common inhabitant of soils, it has been detected in surface waters and laboratory-distilled water[43], and was detected in this study during Phases III and IV.

Gallionella (an iron oxidizer) was shown to exist in 24-hour standing pipe loop water samples during Phases II-IV, with the most significant growth occurring during Phase III and the first portion of Phase IV. Qualitative analysis of growth was indicated by ferrous sulfide agar slants and liquid broth B_{12} medium. It was also probably present in Phase I, since water containing 0.2-0.3 mg/L of iron will almost always contain ferruginous bacteria, but in concentrations too low to be detected.[36] Growth was detected in the ferrous sulfide medium as well as the Vitamin B_{12} medium. Gallionella morphology consists of kidney-shaped or rounded cells on the ends of long "stalks" which are comprised of bundles of fibrils twisted around one another.[30] They may be covered with iron hydroxide precipitates and in older organisms, the cells may detach from the stalk. Gallionella is tentatively identified as the organism in Figures C-40 through C-43).

Desulfovibrio (Figure C-45) increased slightly in 24-hour standing pipe loop water from Phase I through Phase III, which averaged 28 org/100 mL, then gradually began to decrease in Phase IV (Table 18). Barrs' medium and Lewis medium both supported growth of these anaerobic sulfate-reducing bacteria. Desulfovibrio was also present in tubercles, both before and after corrosion treatment. Growth of this organism in tubercles was measured utilizing the Lewis medium; an average of 1.7×10^3 sulfate-reducers per gram of tubercle were detected before corrosion treatment and an average of 2.4×10^3 sulfate-reducers per gram of tubercle were detected after corrosion treatment (as indicated by the MPN method).

Several hypotheses may account for the results observed for *Gallionella* and *Desulfovibrio*. First, *Gallionella* oxidizes ferrous iron to hydrated ferric hydroxide. Table 16 indicates an increase in standing water iron concentrations of approximately 38% over the average untreated standing water iron concentration, thereby providing an increased amount of iron ions for microbial assimilation, and through oxidation, yielding insoluble ferric hydroxide.

As ferrous deposits increased, anaerobic conditions prevailed at the metal surface and the microbial attack became more vigorous as a differential cell was formed.

Second, as the heterotrophic bacteria increased in number after Phase I, oxygen was consumed and anaerobic conditions were created causing environmental conditions to change in favor of the growth of sulfate-reducers such as *Desulfovibrio*.

Sphaerotilus and *Leptothrix*, categorized as sheathed iron bacteria, occur as straight rods within a sheath or free-swimming as single cells, in pairs, or in some species as motile short chains.[30] Encrustation of the sheaths with deposits of iron hydroxide takes place after the cells have left the sheaths. *S. natans* and *L. ochracea* are nearly identical except *S. natans* is unable to oxidize manganous compounds. These two organisms are tentatively identified in Figure C-44.

The importance of detecting these different groups of microorganisms in the pipe loop water was that metabolic activities of a variety of microorganisms could combine to establish microbiotic cycles of essential elements such as carbon, nitrogen, sulfur, iron, phosphorous, etc. A microbial ecosystem could be established in time which could potentially influence the concentrations of numerous constituents in the water. For example, heavy growths of iron bacteria can alter the physiochemical properties of pipe surfaces and serve as nutritional substances for various chemoorganotrophic microorganisms. The metabolic activities of the latter under anaerobic conditions can produce organic acids and hydrogen sulfide, which in turn, can lead to corrosion problems. High *in situ* concentrations of such microorganisms might conceivably alter systems to produce a microenvironment more favorable for the survival and propagation of specific pathogenic bacteria. Consequently, water quality changes may be attributed to microbial activities in water.

Microbial Quality and Chemical Properties of Tubercular Incrustations in the Pipe Loop

Microorganisms which can grow on the surface of a metal in a coherent clump or colony are potentially capable of producing ion or oxygen concentration cells (tubercles). Throughout their growth period, aerobic organisms

Table 18. Results of Bacteriological Analyses of Specific Corrosion-Related Microorganisms

PHASE	Total # H_2O Samples Collected	# of H_2O Samples Analyzed	Desulfovibrio			Gallionella			Filamentous Organisms*		
			Water (org/100 mL)		Tubercles[†]	Water (org/100 mL)		Tubercles[†]	Water (org/100 mL)		Tubercles[†]
			Control	Standard	(org/cm)	Control	Standard	(org/cm)	Control	Standard	(org/cm)
I	13	8	ND	$+^{a,b}$ 10	$+^{a,b}$ (1700)	ND	ND	$+^{a,b}$	ND	$+^{b}$	$+^{b}$
II	9	6	ND	$+^{a,b}$ 24	--	ND	$+^{a,b}$	--	ND	$+^{b}$	--
III	4	4	ND	$+^{a,b}$ 28	--	ND	$+^{a,b}$	--	ND	$+^{b}$	--
IV	9	6	ND	$+^{a,b}$ 18	$+^{a,b}$ (2400)	ND	$+^{a,b}$	$+^{a,b}$	ND	$+^{b}$	$+^{b}$

(continued)

+: Present.
NM: Not measured.
ND: Not detected, i.e., in low concentration.
[a] Detection by direct isolation techniques.
[b] Observed by scanning electron microscopy.
*Possible organisms might include the Sphaerotilus-Leptothrix group and Actinomycetes, etc.
[†] The amount of tubercular incrustations (grams) examined after Phase I and Phase IV was equivalent.

Table 18, Cont.

PHASE	Total # H_2O Samples Collected	# of H_2O Samples Analyzed	*Caulobacter* and *Hyphomicrobium* spp.		
			Water (org/100 mL)		Tubercles[†] (org/cm)
			Control	Standard	
I	13	8	+[b]	+[b]	ND
II	9	6	+[b]	+[b]	--
III	4	4	ND	+[b]	--
IV	9	6	+[b]	+[b]	+[b]

+: Present.
NM: Not measured.
ND: Not detected, i.e., in low concentration.
[a] Detection by direct isolation techniques.
[b] Observed by scanning electron microscopy.
*Possible organisms might include the *Sphaerotilus-Leptothrix* group and *Actinomycetes*, etc.
[†] The amount of tubercular incrustations (grams) examined after Phase I and Phase IV was equivalent.

can absorb oxygen or nutrients from the immediate surroundings of cell clumps. The oxygen (or nutrient) concentration under a cell mass may become reduced as a result of microbial metabolism. A potential may be formed between two areas: the area underneath the cell mass and areas adjacent to the cell mass.

Iron metal would be expected to go into solution at the area underneath the cell mass (anode) and the resultant electrons may be combined with dissolved oxygen and water to form hydroxyl ions at the oxygenated cathodic areas. As long as the concentration cells persists, the corrosion process will also ensue, resulting in localized corrosion, known as "pitting." Figures C-46, C-47, and C-48 show the tubercles as they appeared in the pipe loop. Iron oxide material in exterior layers forms a hard-textured crust on the tubercle surface. In Figure C-47, a tubercle has been removed and the pitting underneath is visible.

The interior of fully-developed tubercles (Figure C-47) is chambered, which according to Stampfl[44] suggests that the tubercle formation proceeds at a non-uniform rate that may be determined by periodic fluctuations in the temperature and chemical composition of the water and perhaps rupture of an exterior crust. Tubercle interiors have a high content of black fluid in association with complex iron oxyhydroxides that crystallize as green rust.

When the surfaces of tubercles from the pipe loop were examined in the scanning electron microscope, they were observed as being coated with a superficial layer of flocculant precipitate (Figures C-49 through C-52). Closer inspection of the precipitate at higher magnification revealed dense microcolonies of various bacteria. Such microcolonies were randomly distributed over the nodule surface, and in some areas the entire nodule surface was covered with a mat-like layer of cells up to several micrometers in thickness. These layers of filaments often acted as a trap for various bacteria, algae, and diatoms (Figures C-53 through C-58). Allen, *et. al.*[45] found colonization of microorganisms and resultant high bacterial populations often occurred on or within protective pipe incrustations.

The most frequently observed microorganisms were filamentous bacteria (Figures C-59 and C-60). They measured approximately 0.5 to 1.0 μm diameter and up to 50 μm in length. The ends of these filaments were often hidden from view by mineral deposits, hence preventing an accurate determination of their true length. Some filaments closely resembled certain members belonging to the *Actinomyces*. Many of the filaments penetrated into openings within the mineral layer, thereby appearing to firmly anchor the cells to the pipe surface.

Gallionella was observed attached to pipe surfaces and often heavily encrusted with mineral deposits. These organisms obtain their energy chemolitotrophically, presumably by enzymatic oxidation of soluble ferrous iron to the ferric state.[27] This process results in the accumulation around the cell and stalk of insoluble ferric salts (principally the hydroxide form). These substances appear in the scanning microscope as small spherical nodules adhering to the stalk surface (Figures C-61 and C-62).

The oxalic acid used in the preparation of some tubercles for SEM clearly dissolved the amorphous iron; however, the granular iron, partially crystallized, was not significantly reduced in number or size (Figure C-63). By these acid treatments, it was possible to demonstrate extracellular layers of fibrous material (Figures C-52, C-53, and C-63 through C-68). The cells were not considerably plasmolyzed by the treatments. However, many cellular structures were still undiscernible, owing to the presence of iron deposition.

Many bacteria attach to solid surfaces in aquatic habitats by a network of extracellular mucopolysaccharide fibrils (i.e., glycocalyx) having a heteropolysaccharide or glycoprotein composition.[46] Such extracellular fibrils can interact specifically with the substratum and have been frequently observed on bacterial surfaces.[47, 48] Extracellular fibrils, similar in appearance to those previously reported by other workers, were evident extruding from some of the bacteria attached to the pipe incrustations (Figures C-62, C-64, and C-69).

Elemental Composition of Tubercles and Pipe Loop Water

Due to the fact that the chemistry of the pipe surface might conceivably strongly influence the ability of certain microorganisms to attach and multiply, the total elemental composition of the encrusting mineral layer was determined by using X-ray energy-dispersive microanalysis (EDAX). Figures C-71, C-73, and C-75 display X-ray energy spectrums of several corresponding pipe incrustations (Figures C-70, C-72, and C-74). Iron was the major cationic element in the tubercles and occurred primarily in the ferrous form. Other predominant elements on the pipe surface included aluminum, silicon, zinc, manganese, and phosphorous. The pattern of distribution of major and trace elements was more or less similar for each tubercle analyzed. The level of sulfur in the tubercles was quite low, suggesting that iron sulfides were not major corrosion products when compared with iron oxides. Accumulations of zinc, manganese, silicon, and phosphorous increased in the outer tubercle crust, while iron appeared to be present in higher concentrations in the tubercle interior (see Figures C-74 and C-75). Figure C-77 displays the decrease in iron after a tubercle (Figure C-76) was subjected to oxalic acid treatment.

The stoichiometry of the elements comprising the mineral layer was quite different from that of the running and control water samples; however, it was similar to that of standing water samples, i.e., while the running and control samples were quite low in concentrations of iron, aluminum, silicon, zinc, manganese, and phosphorous, standing water samples readily showed elevated levels of these same metals (see Table 16 and Figures C-78 through C-85).

Results from this study indicate that bacteria attached to the pipe surface or those bacteria subjected to long residence times (viz., in premise plumbing, where chlorine residuals are often nonexistent) are exposed to concentrations of minerals or organic micronutrients which are significantly different from those found in the constantly flowing distribution water. Such altered concentrations of minerals and nutrients can have a profound effect on the growth kinetics of such microorganisms and their susceptibilities to chlorine residuals (i.e., if chlorine residuals are even present).

Thus, results received in this study from the investigation of various parameters indicated that proliferation of bacterial microcolonies on pipe surfaces and those associated with suspended particulate matter in premise plumbing could lead to a significant underestimation of the total number of viable microorganisms associated with a given water supply, thereby making accurate public health assessments of the water ultimately received by the consumer difficult or impossible to achieve.

The Effects of Treatment Start-Up on Chemical and Physical Parameters

Zinc Leaching --

Figure 38 illustrates the effect of treatment start-up on 24-hour standing zinc concentrations. Upon initial treatment of Tolt water to an average pH of 7.11, zinc levels were reduced by approximately 61%. The pH was then increased to an average of 7.37. This was followed by a small spike in zinc leaching, which quickly dropped to approximately equal reduction levels. Upon full treatment to an average pH of 8.08, zinc levels increased to a final reduction of 50%.

The effect of pH on standing water zinc levels is presented in Figure 39. Figure 39 illustrates that zinc leaching in standing water decreases from pH 6 to approximately pH 7, and increases from pH 7 to pH 8. This data is consistent with the results from the Cedar and Tolt pilot tests. The Cedar tests showed an increase in zinc leaching with treatment, while the Tolt tests showed a decrease with treatment.

The data presented in Figure 39 supports the hypothetical zinc corrosion model detailed in Volume I, Cedar River Water Pilot Plant Study.[8] The model states that under standing water conditions, localized zinc corrosion occurs, creating local pH differences and a nonuniform corrosion film. At cathodic sites, dissolved oxygen is reduced thereby increasing the pH at those sites; and, at anodic sites, the oxidation of zinc decreases the pH. At an initial pH of 8, the passivation film becomes unstable at both the anode and cathode, while at pH 7 the film is only unstable at the anode. The result is then greater zinc leaching at pH 8 than at pH 7.

As was also shown in the Tolt and Cedar pilot tests, zinc corrosion in running water was reduced with treatment. The running water metals results presented in Appendix C demonstrate that increased zinc leaching from pH 7 to pH 8 does not occur in running water. Instead, running water zinc levels were consistently reduced by approximately 72%.

Iron Leaching --

Figure 40 illustrates the effect on treatment of iron leaching from old galvanized pipe. The results indicate an increase of approximately 38% over the average untreated standing water iron concentrations. The increased iron concentrations may be due to the diminished action of the zinc as a sacrificial anode. As the zinc is passivated, its sacrificial effect on iron is

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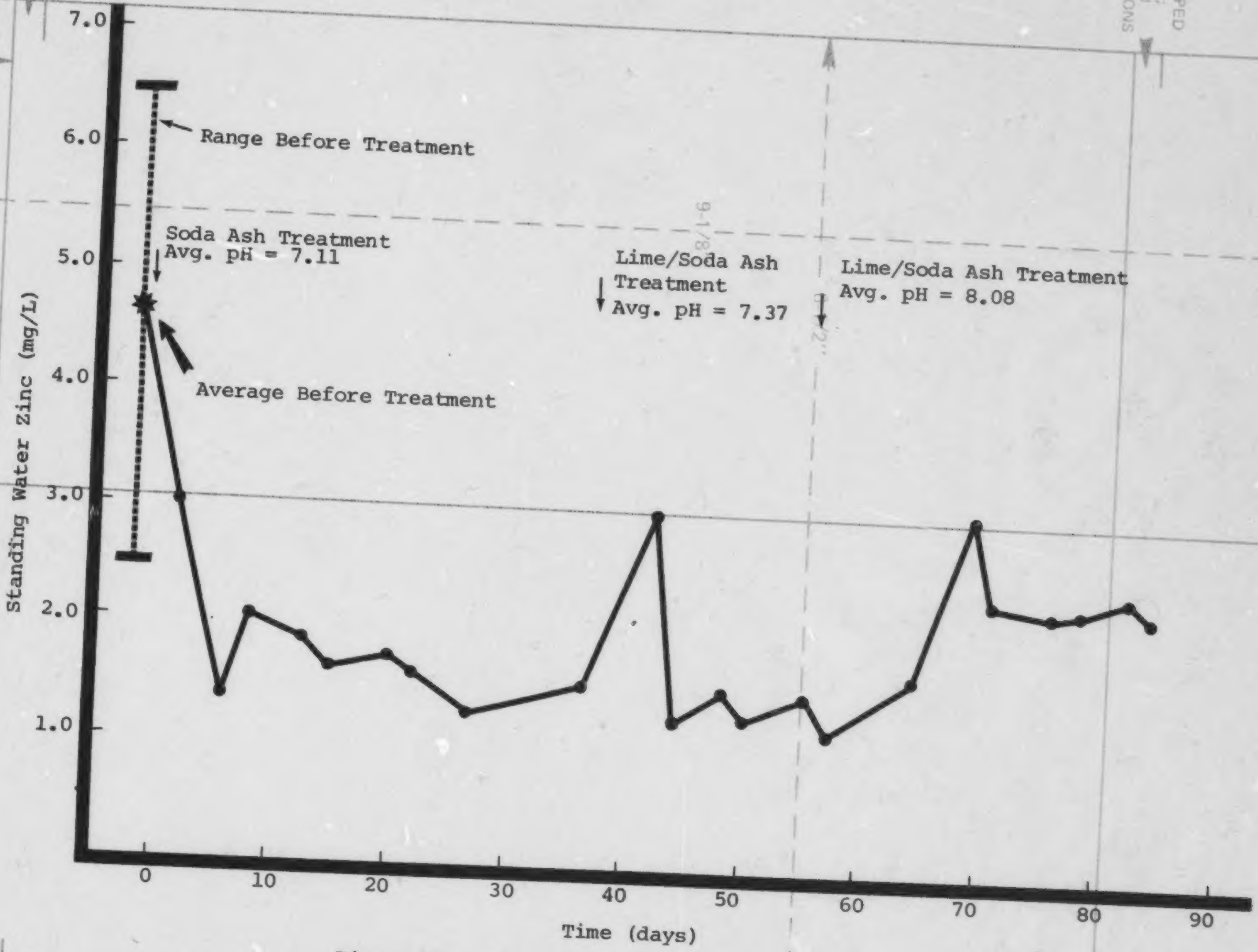

Figure 38. Zinc leaching from galvanized pipe.

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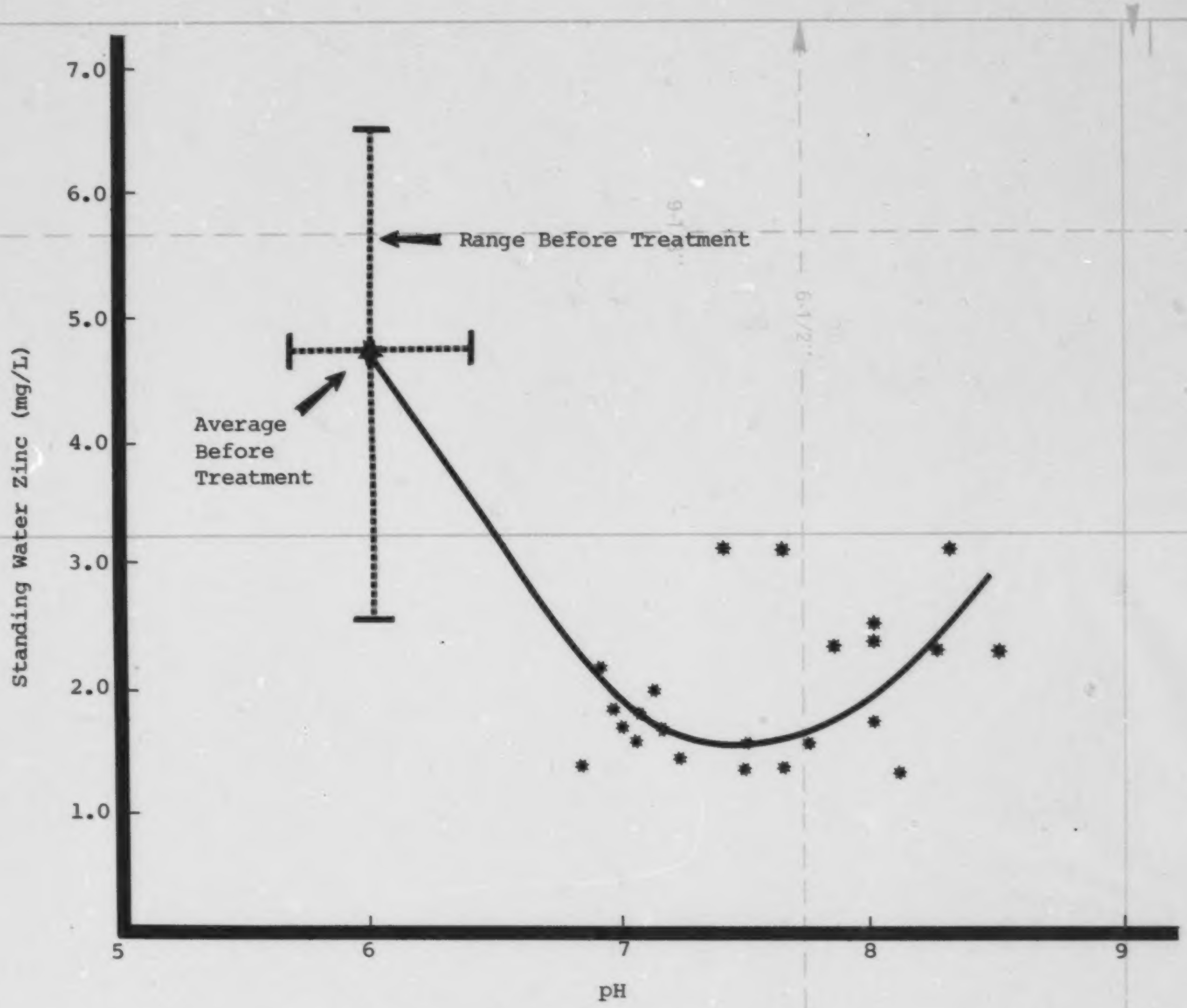

Figure 39. Zinc leaching versus pH.

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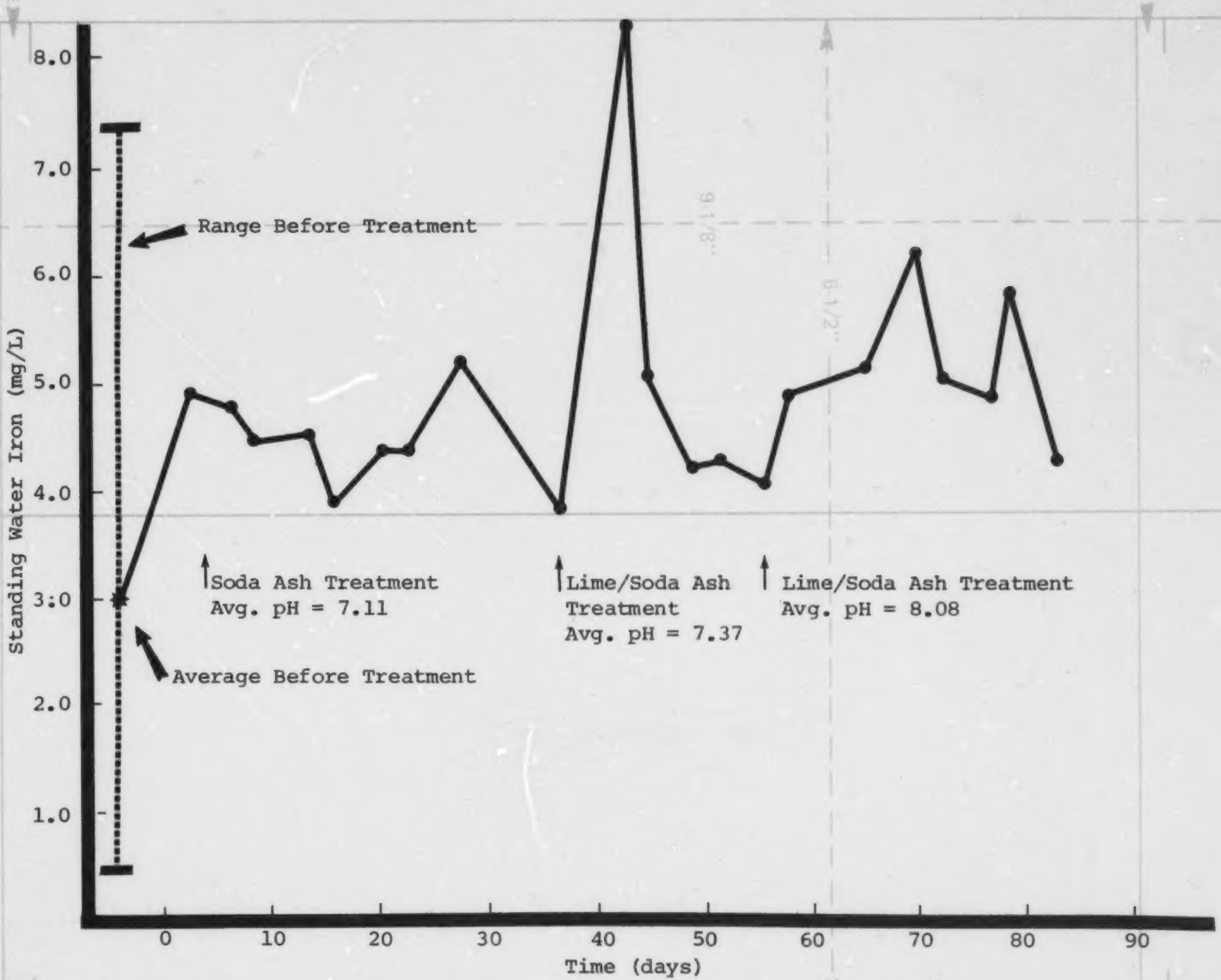

Figure 40. Iron leaching from galvanized pipe.

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diminished thereby allowing iron to more readily corrode.

These tests agree somewhat with earlier tests which demonstrated only very slight corrosion rate reductions for iron. The Tolt pilot plant study showed increases with treatment of 33% and 10% in the weight loss and pitting rates of black steel, respectively. The Cedar pilot plant showed weight loss and pitting rate decreases of only 6% with treatment.

Appendix B presents running water iron leaching results. As with zinc leaching, running water iron levels were substantially reduced with treatment. Average running water iron reductions were approximately 61%.

Lead Leaching --

Lead leaching results are presented in Figure 41. Standing water lead concentrations were reduced by 57% during treatment start-up. Running water lead levels showed no appreciable difference both before and after treatment.

Cadmium Leaching --

Cadmium leaching results are presented in Figure 42. As with the zinc leaching results, standing water cadmium concentrations initially decrease with treatment to pH 7, but then increase from pH 7 to pH 8. The Cedar[8] and Tolt[7] pilot studies also showed similarities between zinc and cadmium leaching.

The same type of corrosion mechanism is involved in cadmium corrosion as in zinc corrosion. Pourbaix15 diagrams for cadmium and zinc are very similar, showing unstable passivation films at low and high pH's for both metals.

Treated running water cadmium concentrations showed no appreciable change over untreated samples.

Copper Leaching --

Although very little copper pipe was contained in the building plumbing system, copper concentrations increased in standing water compared to running water copper levels from Phase I through Phase IV. Copper leaching results are presented in Figure 43. Standing water copper concentrations were reduced by 53% from Phase II through Phase IV. Running water copper levels showed no appreciable difference before and after treatment.

Apparent Color --

The apparent color of standing and running water responded similarly to treatment as did standing and running iron concentrations. Figure 44 demonstrates that standing water color levels increased by an average 23% with treatment over the untreated standing water conditions. The running water color levels remained essentially constant throughout treatment, with no deviation from the untreated running water level (Figure 45). Most of the apparent color is due to the iron concentration in the sample.

Turbidity --

Figure 46 illustrates the effects of treatment start-up on 24-hour standing water turbidity levels. During initial treatment of Tolt water to

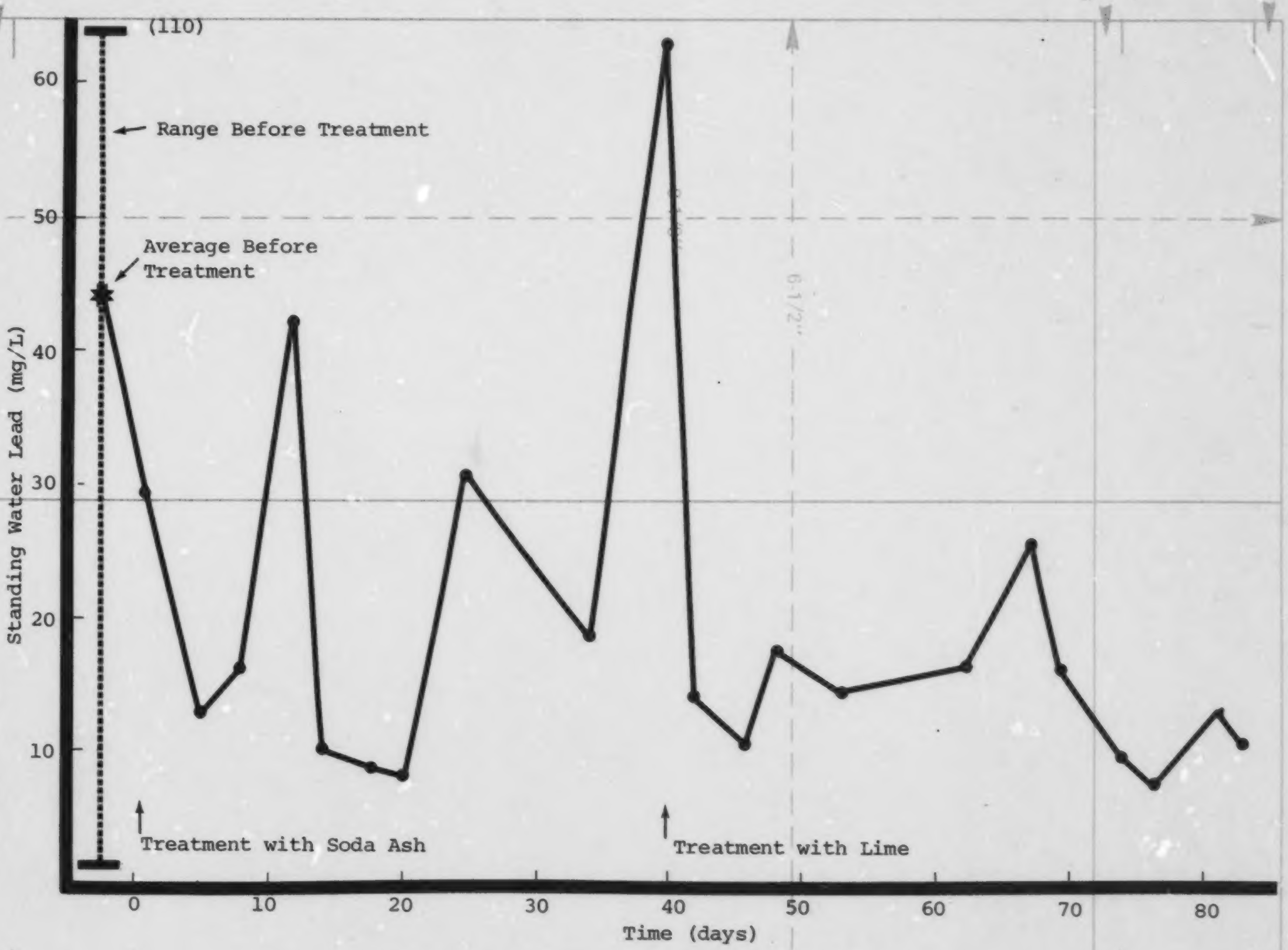

Figure 41 . Lead leaching from galvanized pipe.

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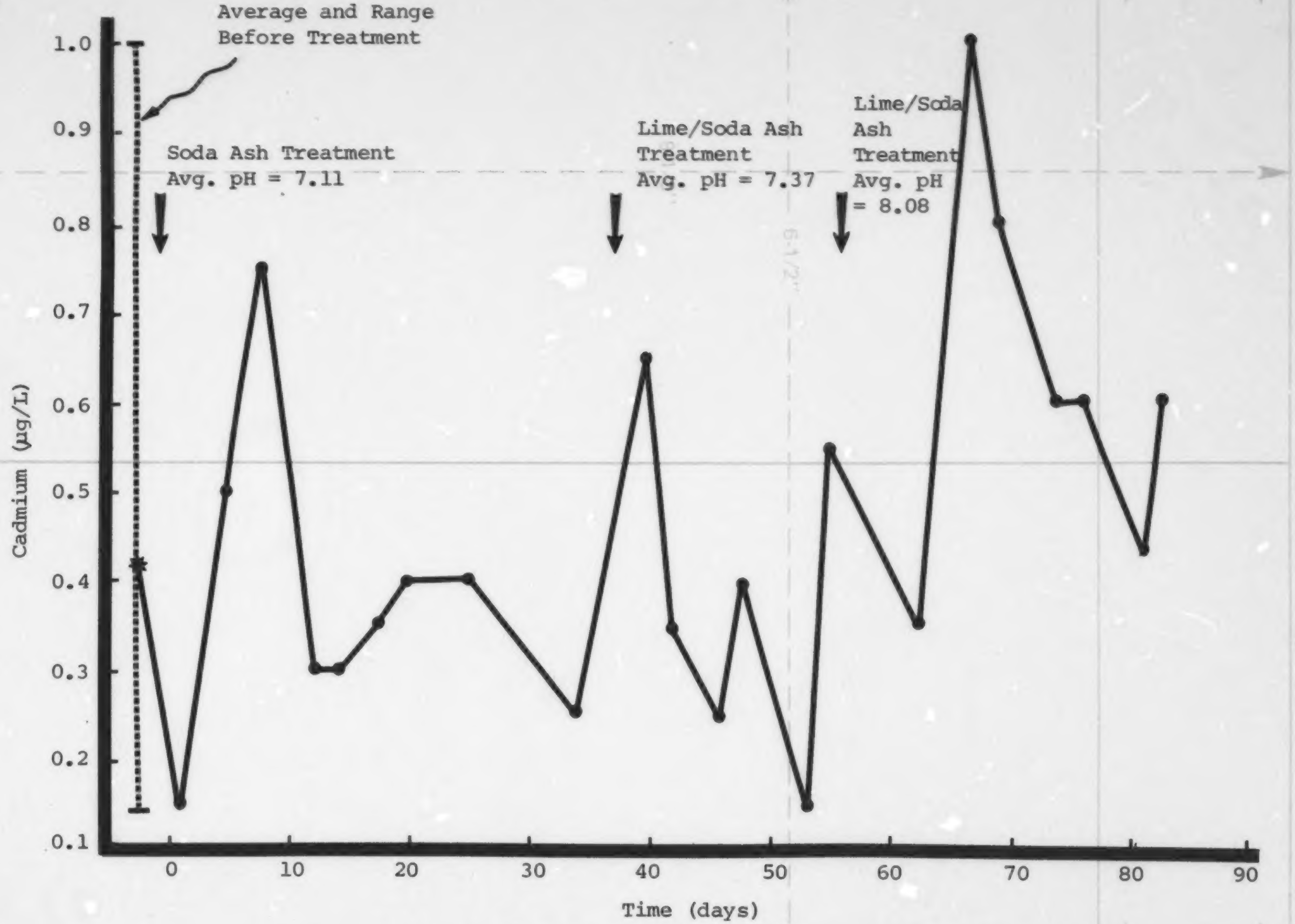

Figure 42. Cadmium leaching from galvanized pipe.

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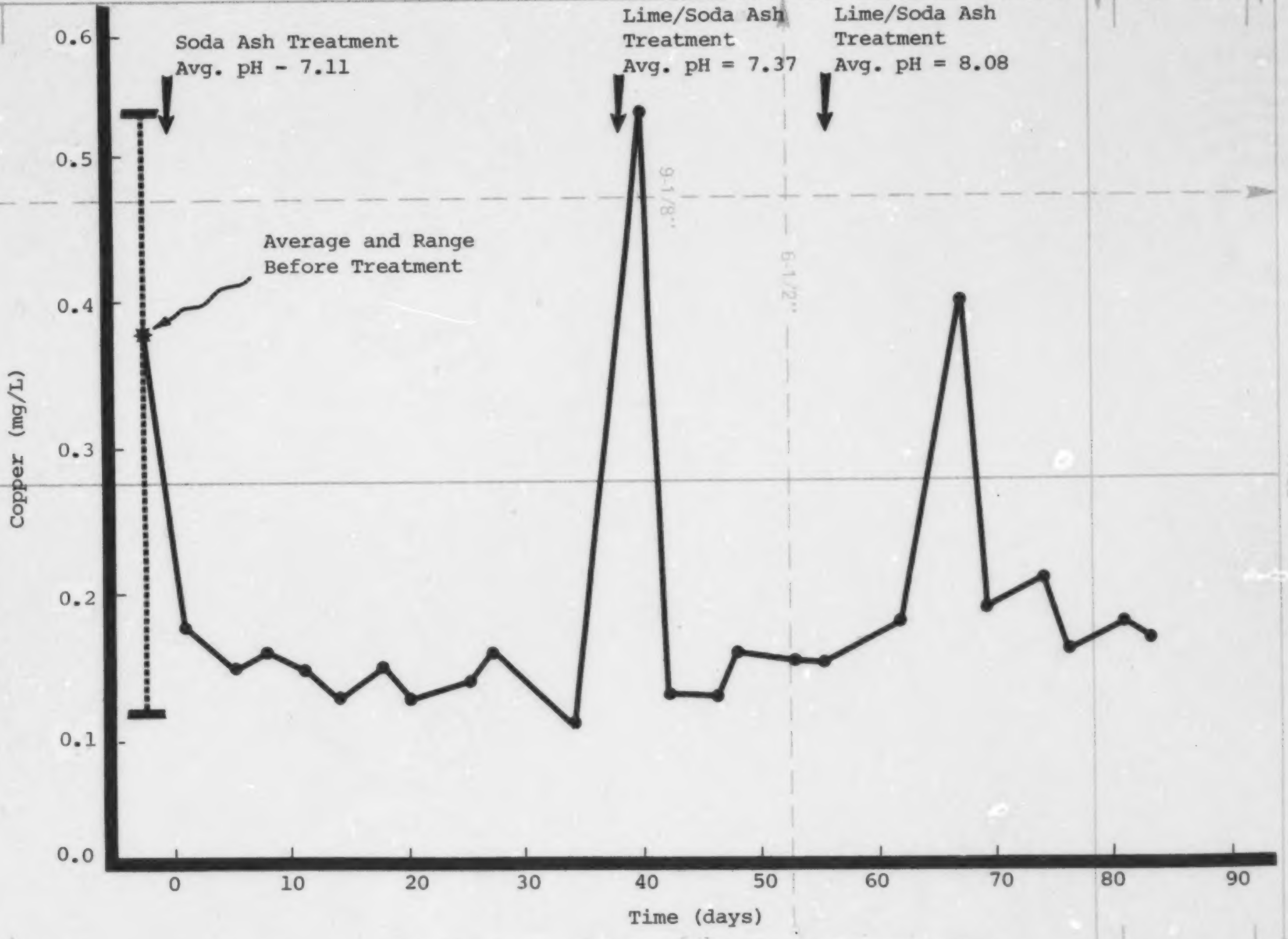

Figure 43. Copper leaching from galvanized pipe.

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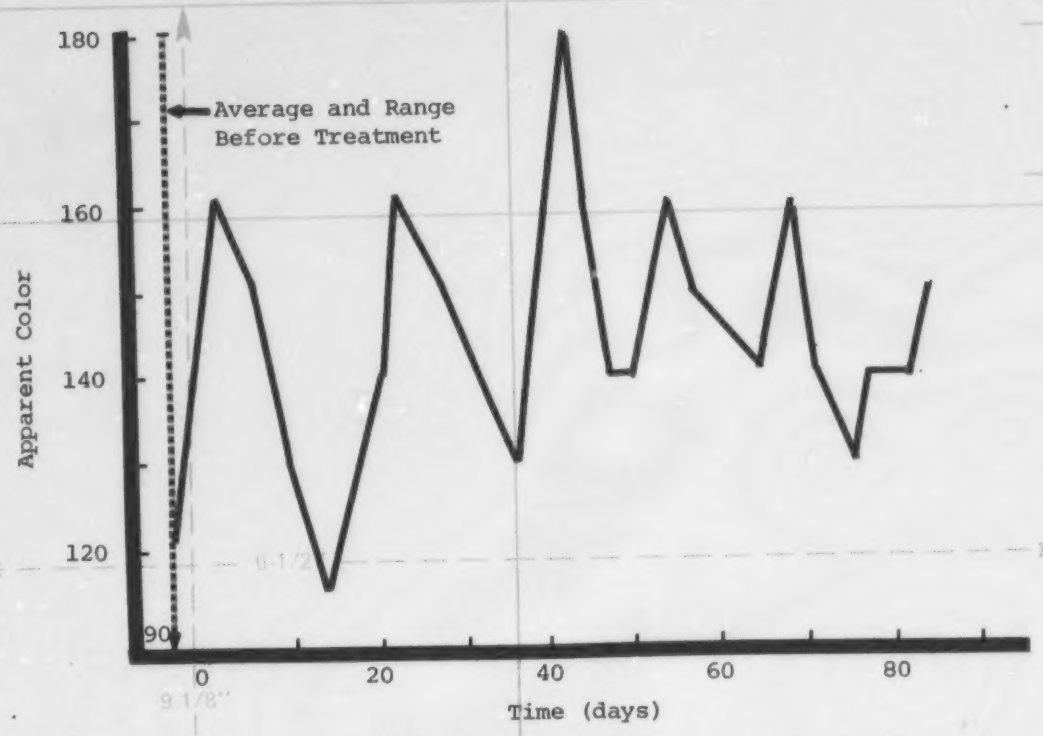

Figure 44. Standing water color versus time.

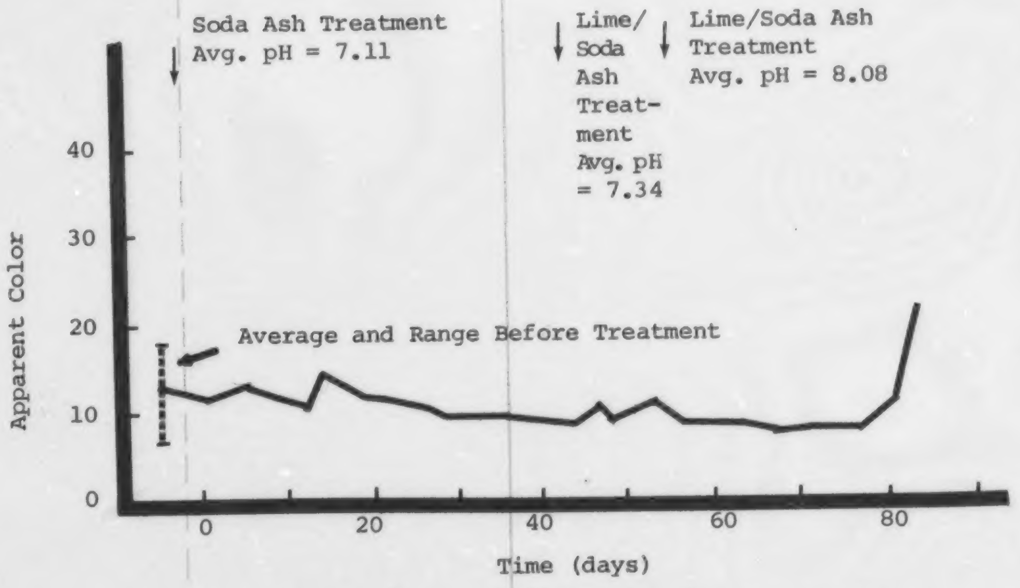

Figure 45. Running water color versus time.

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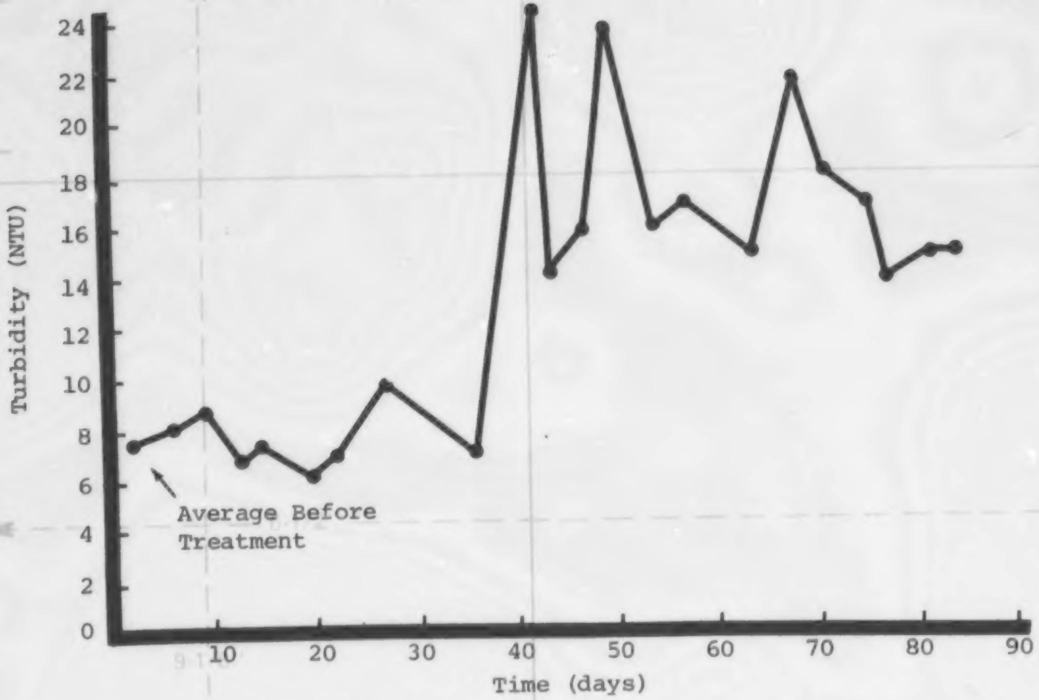

Figure 46. Standing water turbidity versus time.

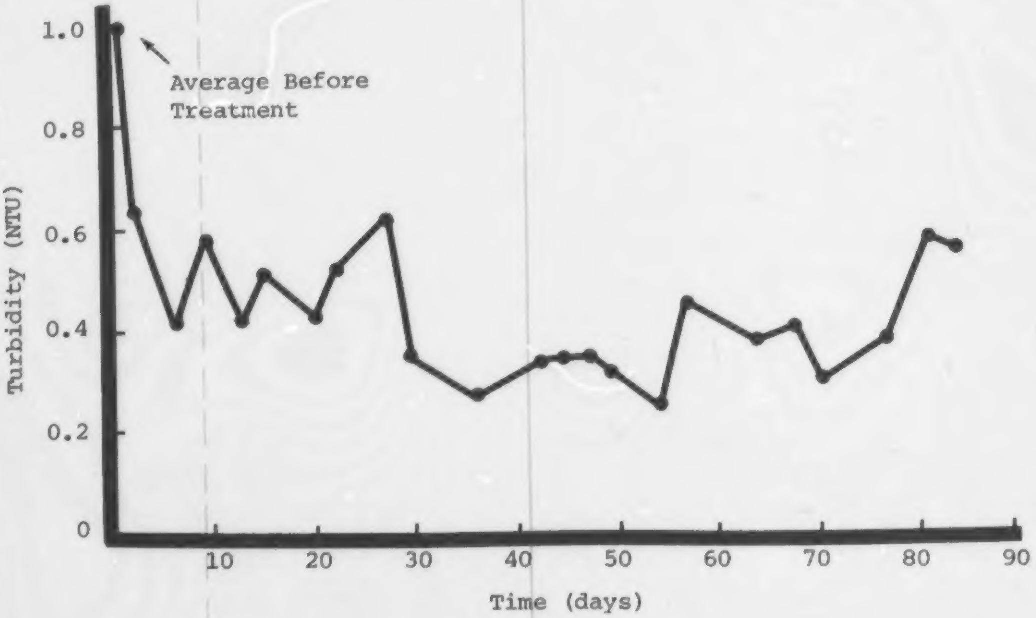

Figure 47. Running water turbidity versus time.

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an average pH of 7.11 (Phase II), turbidity levels increased slightly over those of untreated water, showing an average increase of 22%. With continued treatment to pH levels of 7.37 and 8.08, standing water turbidity increased by 57%.

Figure 47 illustrates the effects of treatment start-up on running water turbidity levels. All three pH levels displayed similar readings, with a mean turbidity reading of 0.51 NTU. This is much less than the 24-hour standing water mean turbidity reading of 15.0 NTU.

Total Suspended Solids --

Figure 48 presents the effects of treatment start-up on total suspended solids standing water concentrations. Suspended solids concentrations were reduced by approximately 20% in standing water throughout the treatment start-up. This indicates that sloughing off of iron tubercles in pipes will not occur during full-scale treatment start-up.

Headloss --

Headloss was used as an indicator of tuberculation build-up in the pipe loop system. Figure 49 shows the increase in headloss with time both before and after treatment. The dashed lines in this figure represent the least squares fit of the data points before and after treatment, respectively. The slope of these lines is equal to the average rate of increase in headloss. Figure 49 demonstrates this rate was decreased by approximately 55% with treatment, which indicates that premature plugging of galvanized pipe will not occur because of corrosion treatment.

Summary of Onsite Tests

Corrosion treatment start-up should have both negative and beneficial effects on water quality. Bacteriological results presented in the previous sections indicate that standing water quality in old galvanized systems will demonstrate increased bacterial populations with treatment start-up. Opportunistic pathogens and coliform antagonists, such as *Pseudomonas* spp. and *Flavobacterium* spp., were some of the bacteria showing the greatest increase with treatment start-up. Ferruginous bacteria, specifically *Gallionella* spp., will most likely increase with treatment start-up. In addition, sulfate-reducers, e.g., *Desulfovibrio*, should increase in number, both in the standing water as in the pipe incrustations. Some standing water quality parameters, such as iron concentration, color, and turbidity, increased with treatment and may present a problem during treatment start-up. It should be noted that this work represents a "worst case" condition and that the actual response of a plumbing system to corrosion treatment start-up will vary depending on the type, age, and condition of the plumbing. It is expected, however, that running water quality will be improved during treatment start-up. Also, premature plugging of pipes will not occur because of treatment.

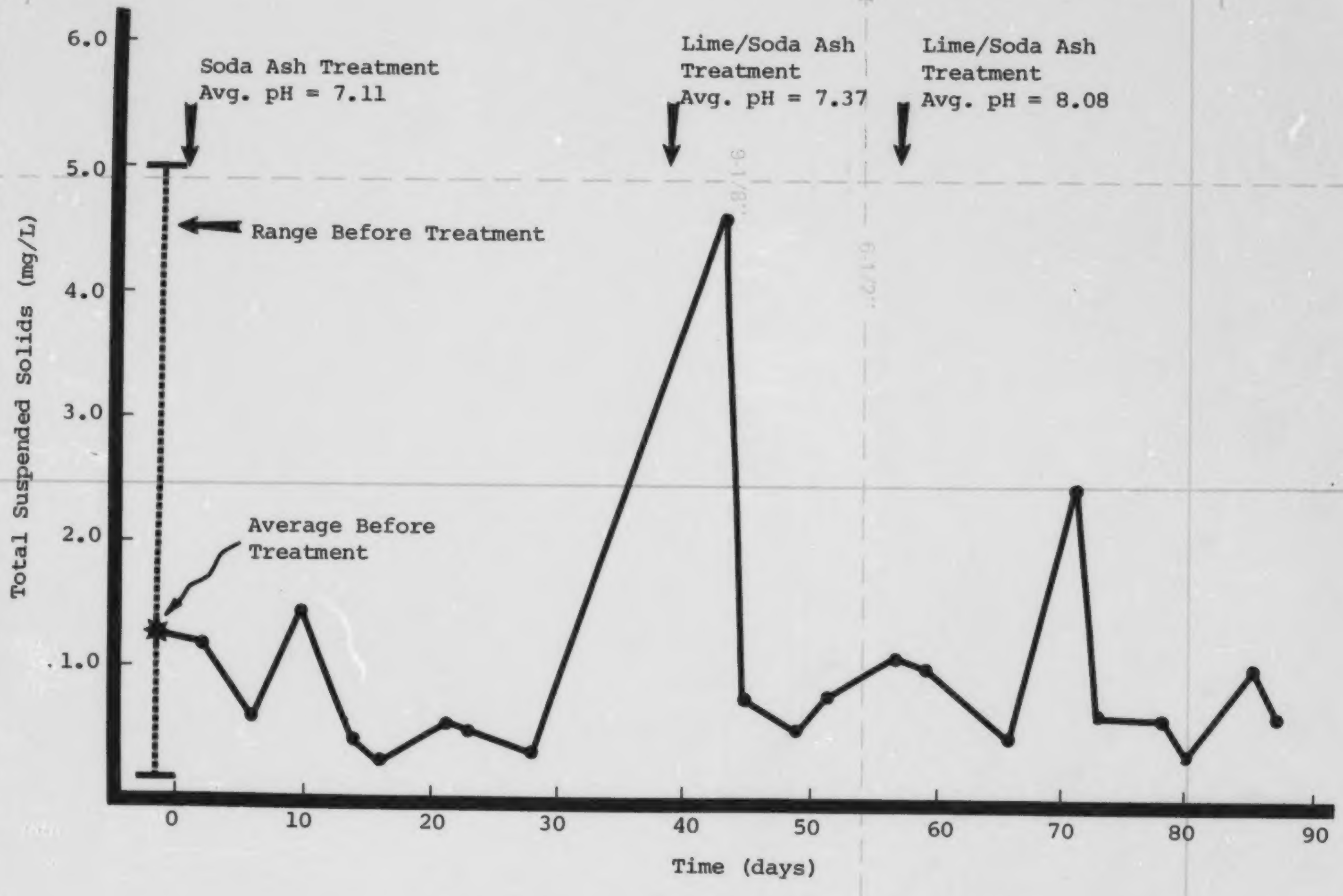

Figure 48. Total suspended solids versus time.

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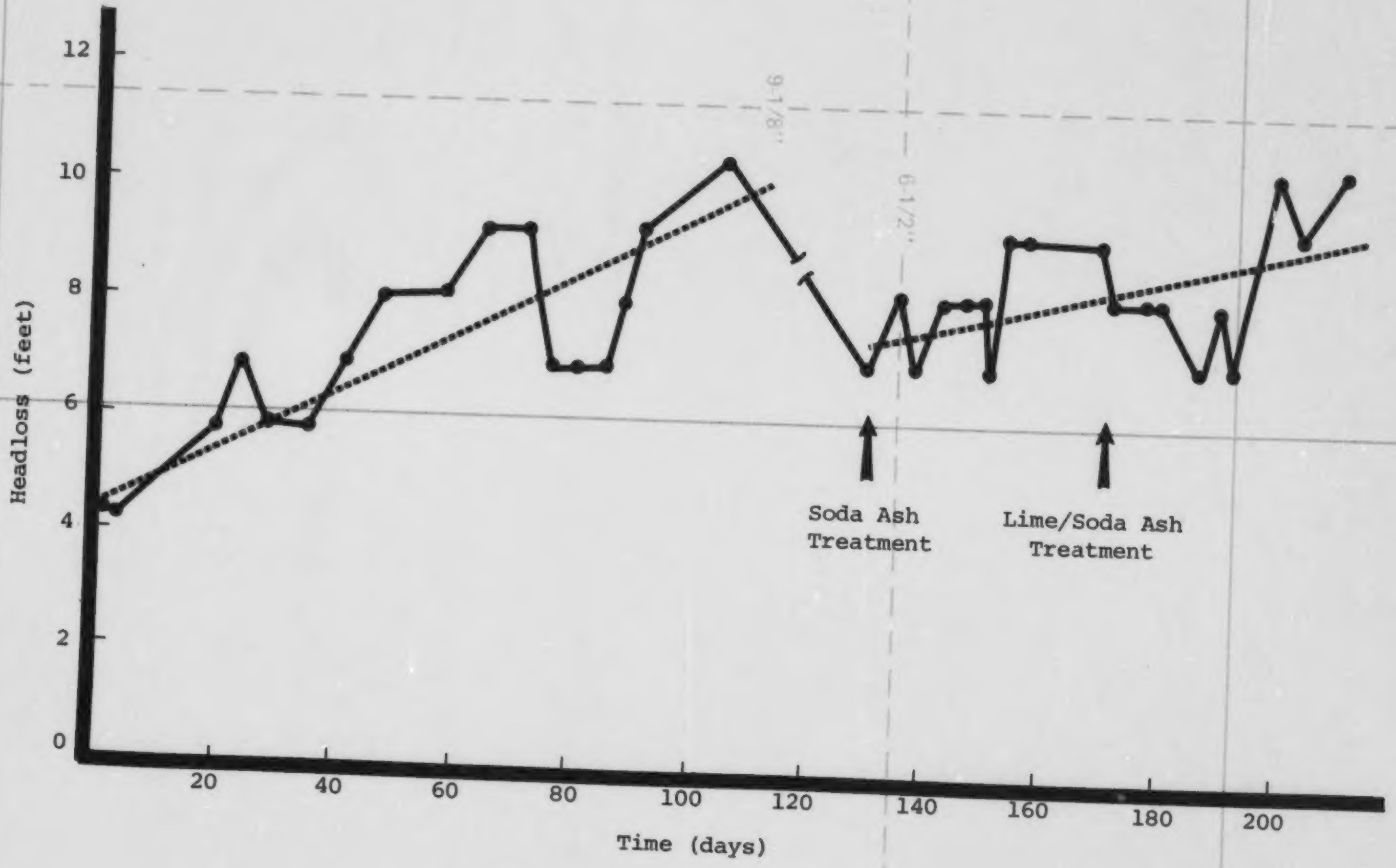

Figure 49. Headloss versus time.

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APPENDIX A

SELECTIVE ENRICHMENT CULTURE MEDIA

Media		Organism(s) Isolated
Reinforced Baars' Medium (Postgate's Medium B):[1]	pH 7.0, [$K_2 \cdot HPO_4$, 0.05%; NH_4Cl, 0.1%; $CaSO_4$, 0.1%; $MgSO_4 \cdot 7\ H_2O$, 0.2%; $Fe(NH_4)_2(SO_4)_2 \cdot 6\ H_2O$, 0.05%; sodium thioglycollate, 0.06%; ascorbic acid, 0.1%; yeast extract, 0.1% and sodium lactate, 0.35%] (freshly prepared on the day of experiment). The culture medium was added to a 60 ml bottle containing 30 ml of sample which was then filled to the brim and stoppered so as to exclude air. Incubation was at 28-30°C for 4-21 days.	Sulfate-reducers (Desulfovibrio)
Lewis Medium:[2,5,6]	pH 7.0, [Na_2SO_4, 0.15%; $K_2 \cdot HPO_4$, 0.05%; $Fe(NH_4)_2(SO_4)_2 \cdot 6\ H_2O$, 0.033%; $CaCl_2$, 0.01%; beef extract, 0.1%; peptone, 0.2%; sodium lactate, 0.35%; sodium ascorbate, 0.01% and $MgSO_4 \cdot 7\ H_2O$, 0.2%] (freshly prepared on the day of experiment). For pipe loop water, qualitative isolation of sulfate-reducers included streaking cultures on the Lewis medium solidified with 1.5% agar followed by incubation at 28-30°C in an anaerobic jar, and incubation of pour plate cultures at 28-30°C in an anaerobic jar, both under H_2. For quantitative enumeration of sulfate-reducers in tubercles and pipe loop water, the MPN series was used.	Sulfate-reducers (Desulfovibrio)
Ferrous Sulfide Agar:[3,5]	pH 6.6, [NH_4Cl, 0.1%; $K_2 \cdot HPO_4$, 0.05%; $MgSO_4 \cdot 7\ H_2O$, 0.02%; $CaCl_2$, 0.01% and $Fe(NH_4)_2(SO_4)_2 \cdot 6\ H_2O$, 0.5%]. Inoculated with a 1-5 ml suspension of Gallionella deposit. Incubated at 25°C for 18-36 hours.	Gallionella

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SELECTIVE ENRICHMENT CULTURE MEDIA, Cont.

Media		Organism(s) Isolated
Liquid Broth B_{12} Medium:[4]	pH 7.0; [glucose, 0.2%; peptone, 0.2%, $MgSO_4 \cdot 7H_2O$, 0.2%; $CaCl_2$, 0.05%; $FeCl_2$, 0.01%; vitamin B_{12}, 0.01%]. A 10-ml sample was mixed with 10 ml of broth or 200 ml of sample was filtered through a 0.45 µm membrane filter. Incubated at 26°C for 48 hours. For identification of individual cells, the electron microscope was used.	Gallionella Sphaerotilus Caulobacter
M-PA:[5]	pH 7.1; [L-lysine HCl, 0.5%; NaCl, 0.5%; yeast extract, 0.2%; xylose, 0.125%; sucrose, 0.125%; lactose, 0.125%; phenol red, 0.008%; $Na_2(SO_4)_2$, 0.50%; $MgSO_4 \cdot 7H_2O$, 0.15%; and ferric ammonium citrate, 0.08%]. (In an agar base, the following antibiotics were added: sulfapyridine, kanamycin, naladixic acid; and actidione)(membrane filter procedure used; plates incubated at 41.5 ± 0.5°C for 48 hours). For confirmation of typical and atypical colonies, milk agar was used and incubated at 35°C for 24 hours.	Pseudomonas aeruginosa

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APPENDIX B

QUALITY ASSURANCE DATA FOR ON-SITE TREATMENT TEST

Table B-1. Periodic Quality Control Analysis

Date 1981	Flow L/min	Untreated pH	Untreated Alk. mg/L $CaCO_3$	Untreated Cond. µmhos	Untreated Ca mg/L	Untreated Na mg/L	Treated pH	Treated Alk. mg/L $CaCO_3$	Treated Cond. µmhos	Treated Ca mg/L	Treated Na mg/L	Treated Na_2CO_3 mg/L	Treated CaO Dosage mg/L
1/6	10.5												
1/9	10.5												
1/12	10.5												
1/14			2.00	26.4									
1/15	10.5												
1/26	3.1												
1/27	10.2												
1/30	10.5												
2/3	10.5												
2/4	10.3		1.6	26.8									
2/10	8.8												
2/11	10.5		1.6	27.6									
2/18	10.5												
2/24	8.2												
2/25	10.5		2.0	25.4									
3/3	9.9												
3/4	10.3		2.2	26.7									
3/10	10.3												
3/11	10.5		1.68	25.3									
3/17	8.8												
3/18	10.9		0.8	23.8									
3/23	11.3												
3/24	10.6		0.9	24.0									
3/26	10.9		0.8	24.0									
3/31	10.2		3.4	24.6									
4/3	10.8		2.2	26.0									
4/6	10.5												
4/7	10.1		1.2	26.8									
4/13	10.5												
4/20	8.6												

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APPENDIX B

QUALITY ASSURANCE DATA FOR ON-SITE TREATMENT TEST

Table B-1, Cont.

		Untreated					Treated						
Date 1981	Flow L/min	pH	Alk. mg/L $CaCO_3$	Cond. µmhos	Ca mg/L	Na mg/L	pH	Alk. mg/L $CaCO_3$	Cond. µmhos	Ca mg/L	Na mg/L	Na_2CO_3 mg/L	CaO Dosage mg/L
5/13*	9.9						7.00	8.8	46.5			7.12	
5/14	9.6		1.8	27.7			7.49	13.2	43.7			12.5	
5/15	10.8		1.6	32.8			7.33	10.2	42.0			9.12	
5/17	11.0						7.40						
5/18	11.0						7.41						
5/19	10.5		1.6	27.4			7.42	8.6	42.0			7.42	
5/20	10.5						7.35						
5/21	10.5	6.15	2.2	27.0			7.41	9.4	39.0			7.63	
5/22	10.2	6.11	2.0	26.3			6.92	8.0	35.2			6.36	
5/25	10.2	6.10					7.12						
5/26	9.9	6.25	2.2	25.4			6.97	13.8	36.5			12.3	
5/27	19.5						7.15						
5/28	11.0	6.12	1.8	26.3			6.90	8.2	35.8			6.78	
5/29	11.0	6.15	1.4	25.2			7.01	8.2	34.9			7.21	
5/30	10.8						7.00						
6/1	10.6	6.05	2.2	28.5			6.95	8.6	35.6			6.78	
6/2	9.9	6.14	2.0	25.3			6.95	9.2	34.8			7.63	
6/3	9.6		2.0	26.0			6.93	9.2	35.6			7.63	
6/4	10.2	6.30	2.0	25.4			6.92	8.8	34.8			7.21	
6/5	11.0	6.11	2.4	24.8			6.89	10.2	37.4			8.26	
6/6	10.8	6.25					7.39						
6/8	10.8	6/20	2.0	25.8			6.85	10.6	40.3			9.12	
6/9	10.8	6.28	2.0	25.3			7.10	9.6	60.0			8.01	
6/10	11.0	6.32					7.20						
6/11	10.6	6.30	3.1	27.1			7.23	11.2	39.1			8.59	
6/12	10.2	6.20	2.2	25.9			7.24	11.2	49.7			9.54	
6/14	9.9	6.15					6.98						
6/15	10.1	6.10					7.00						
6/16	10.2	6.13	2.8	25.9			7.10	9.8	40.5			7.42	
6/17	24.9	6.15	2.2	23.9			7.05	10.8	36.0			9.12	

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APPENDIX B

QUALITY ASSURANCE DATA FOR ON-SITE TREATMENT TEST

Table B-1, Cont.

Date 1981	Flow L/min	Untreated pH	Untreated Alk. mg/L $CaCO_3$	Untreated Cond. µmhos	Untreated Ca mg/L	Untreated Na mg/L	Treated pH	Treated Alk. mg/L $CaCO_3$	Treated Cond. µmhos	Treated Ca mg/L	Treated Na mg/L	Na_2CO_3 mg/L	CaO Dosage mg/L
6/18	10.5	5.98	2.0	24.0			6.95	9.6	40.1			8.01	
6/19	10.5	6.12	2.2	25.3			7.05	9.0	37.4			7.21	
6/21	10.8	6.35					7.10						
6/22†	10.8	6.30					7.60						
6/23	10.8	6.20	2.0	25.0	6.0		7.63	8.2	34.0	10.2			2.35
6/24	10.5	6.10	1.8	26.1	8.0	0.99	7.40	7.6	35.6	11.0	2.62	3.75	1.68
6/25	21.5	6.10	2.0	27.9	7.0		7.50	7.6	37.5	9.6			1.46
6/26	10.5	6.10	2.0	26.4	8.0	0.94	7.10	7.6	36.4	9.2	2.63		0.67
6/28	19.3	6.10					7.23						
6/29	10.2		2.2	27.4	6.4		7.46	7.2	37.6	10.4			2.24
6/30	10.8		1.8	26.7	7.0	0.91	7.48	8.6	37.6	9.4	2.42	3.29	1.34
7/1	10.9	6.13	2.0	26.6	6.8		7.33	8.2	37.4	10.4			2.02
7/2	10.2		1.8	27.6	7.0	1.10	7.59	8.2	39.3	10.8	2.50	3.22	2.13
7/6	9.3	6.15					6.74						
7/7†	9.9		2.0	26.0	7.0	0.95	8.38	11.8	40.9	9.2	2.25	2.99	1.23
7/8	9.9	6.15	1.8	25.3	6.8		7.77	11.0	40.3	9.0			1.23
7/9	11.0	6.20	1.6	27.7	6.8	0.97	8.00	11.0	39.6	9.2	3.60	6.05	1.34
7/10	9.6	6.40					7.47						
7/12	10.3	6.42					7.52						
7/13	10.5	6.40	1.8	26.4	7.0		7.19	9.8	39.3	9.8			1.12
7/14	10.2	6.40	2.0	25.3	7.0		8.60	13.2	13.4	9.2			1.23
7/15	10.5	5.69	1.6	27.2	6.8		8.02	11.0	41.1	9.2			1.34
7/16	10.5	6.15	1.6	26.2	7.0		8.21	11.0	40.6	10.0	4.18	7.52	1.68
7/17	10.3	6.22	2.2	27.1	7.0		8.30	10.8	44.1	10.0			1.68
7/20	10.6	6.20	1.2	26.2	6.8		8.18	11.4	41.7	9.6			1.57
7/21	10.2	6.15	2.2	25.4	7.0	1.04	8.05	11.0	41.6	10.0	3.75	6.23	1.68
7/22	10.2	5.82	1.8	26.6	7.2		8.00	13.0	41.6	10.2			1.68
7/23	11.0	6.30	1.8	25.7	6.6	0.91	8.10	10.8	39.6	10.2	4.03	7.18	2.02
7/24	10.5	6.30					8.32						
7/26	9.8	6.30					8.04						

(continued)

APPENDIX B

QUALITY ASSURANCE DATA FOR ON-SITE TREATMENT TEST

Table B-1, Cont.

		Untreated					Treated						
Date 1981	Flow L/min	pH	Alk. mg/L $CaCO_3$	Cond. µmhos	Ca mg/L	Na mg/L	pH	Alk. mg/L $CaCO_3$	Cond. µmhos	Ca mg/L	Na mg/L	Na_2CO_3 mg/L	CaO Dosage mg/L
7/27	10.8	6.35	1.4	26.7	6.6		8.50	11.4	38.8	9.2			1.46
7/28	10.5	6.19	2.4	25.2	6.4	0.91	8.02	11.6	40.7	8.8	3.56	6.10	1.34
7/29	9.9	6.65	2.8	24.8			8.25	12.4	39.0				
7/30	10.5	6.30	2.0	24.3	6.4	0.93	8.10	12.0	39.2	9.4	3.45	5.80	1.68
7/31	10.5	6.45					8.35						
8/2	10.2	6.45	2.2	26.2			8.40	10.8	40.5				
8/3	10.3	5.95	2.2	25.2			7.99	10.8	39.0				
8/4	10.5	5.90	2.2	25.7	7.0	0.98	8.30	11.2	39.2	10.0	3.91	6.74	1.68
8/5			1.8	26.2	7.0	1.01		11.4	39.1	10.0	4.06	7.02	1.68

Table B-2. Flow Induced Chemical Feed Fluctuations

Flow L/min	pH	Cond. µmhos	Alk. mg/L $CaCO_3$	Ca mg/L $CaCO_3$	Na mg/L	CaO Feed mg/L	Na_2CO_3 Feed mg/L
Control	5.95	26.2	1.8	6.6	0.92	----	----
3.79	7.87	39.4	11.8	8.6	4.32	1.12	7.82
7.5	7.58	39.8	11.4	8.8	4.42	1.23	8.05
11.0	7.85	39.1	11.4	8.6	4.26	1.12	7.68
11.0	8.05	40.7	11.6	9.6	4.35	1.68	7.89
11.0	8.05	40.7	11.2	9.8	4.36	1.79	7.91
11.0	7.85	40.6	11.6	9.0	4.31	1.34	7.80
15.1	8.09	42.2	11.2	9.2	4.38	1.46	7.96
18.9	7.78	40.6	11.6	9.0	4.42	1.34	8.05
22.7	7.85	41.1	11.2	9.6	4.30	1.68	7.77
31.0	7.55	40.7	11.6	8.8	4.37	1.23	7.94
38.6	7.89	41.3	11.0	9.6	4.36	1.68	7.91
46.2	7.71	40.7	11.6	9.2	4.42	1.46	8.05
56.8	7.67	41.1	11.4	9.0	4.54	1.34	8.33

APPENDIX C

SCANNING ELECTRON MICROSCOPY ANALYSIS

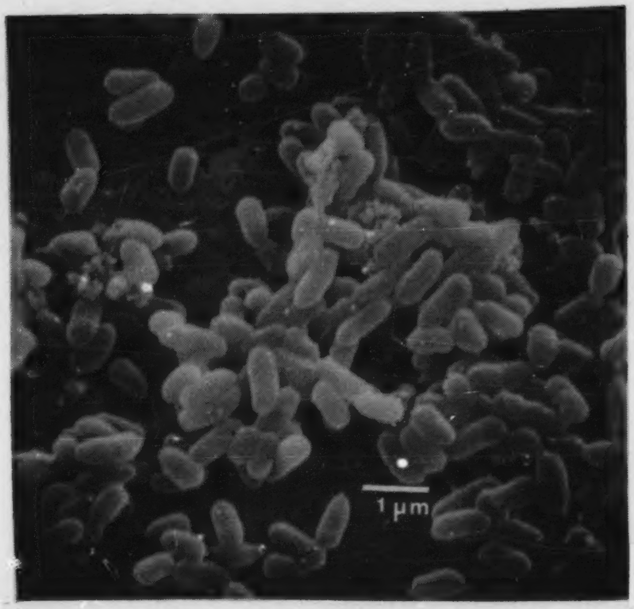

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Figure C-1. Gram negative coccobacilli, isolated from pipe loop water. Yellow-pigmented when grown on standard plate count medium (9,500 X)

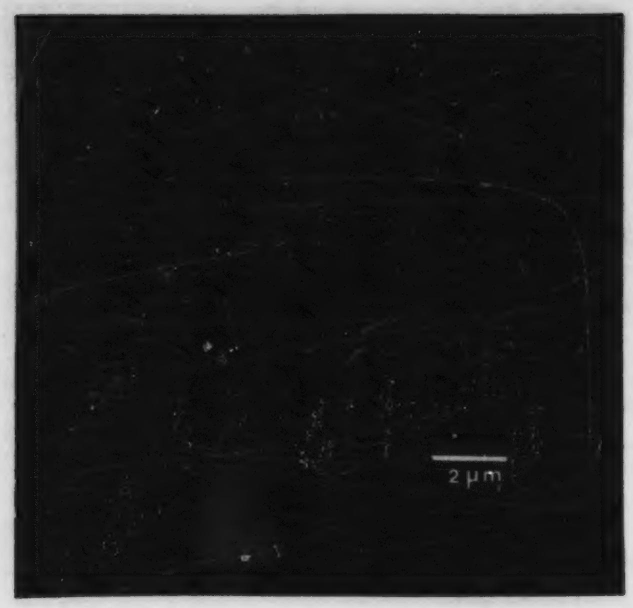

Figure C-2. Gram positive cocci, isolated from pipe loop water. Yellow-pigmented when grown on standard plate count medium (5,220 X).

Figure C-3. Gram negative rods, isolated from pipe loop water. Non-pigmented when grown on standard plate count medium (2,200 X).

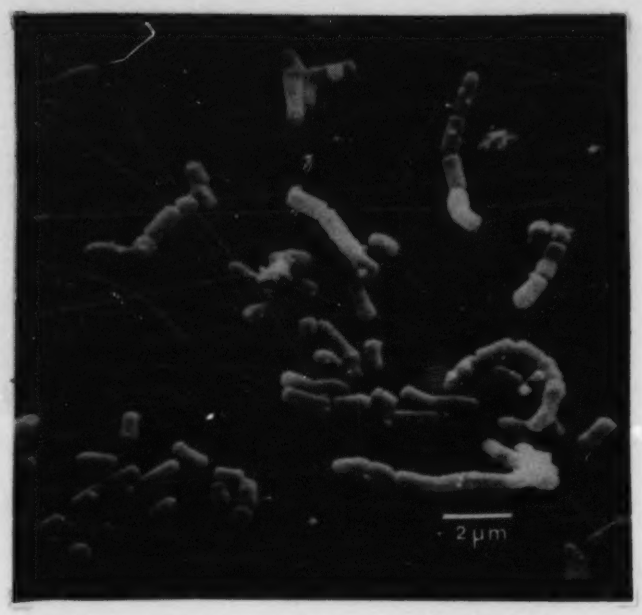

Figure C-4. Gram negative rod-shaped to coccoid, isolated from pipe loop water. Non-pigmented when grown on standard plate count medium (5,010 X).

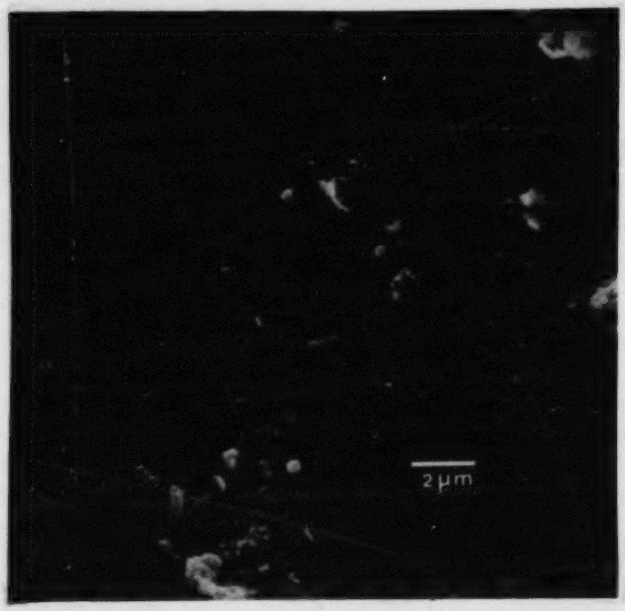

Figure C-5. Phase I: 50 ml of 24-hour standing water filtered through a 0.2 μm Nuclepore filter (4,750 X).

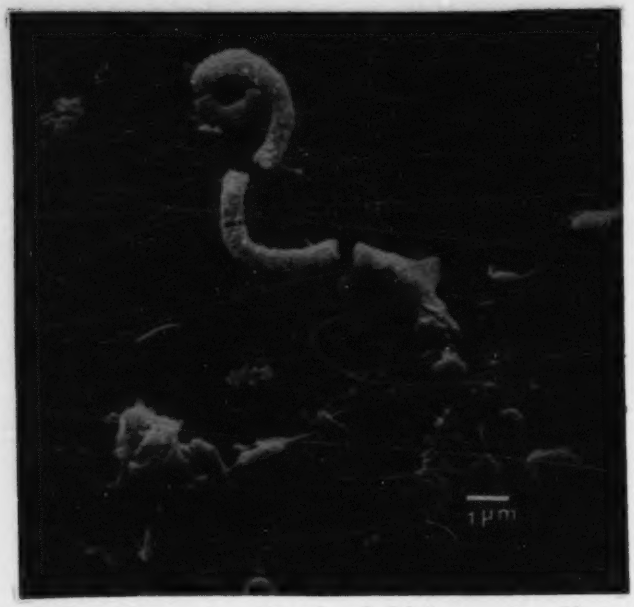

Figure C-6. Phase I: 50 ml of 24-hour standing water. Note filaments, particulate matter, and disassembled spiral-shaped bacterium (5,700 X).

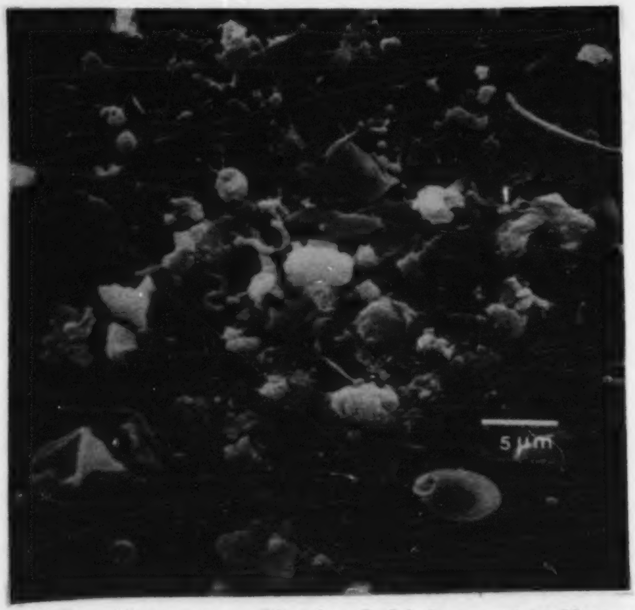

Figure C-7. Phase I: 50 ml of 24-hour standing water. Note spiral-shaped bacteria and organic debris (2,260 X).

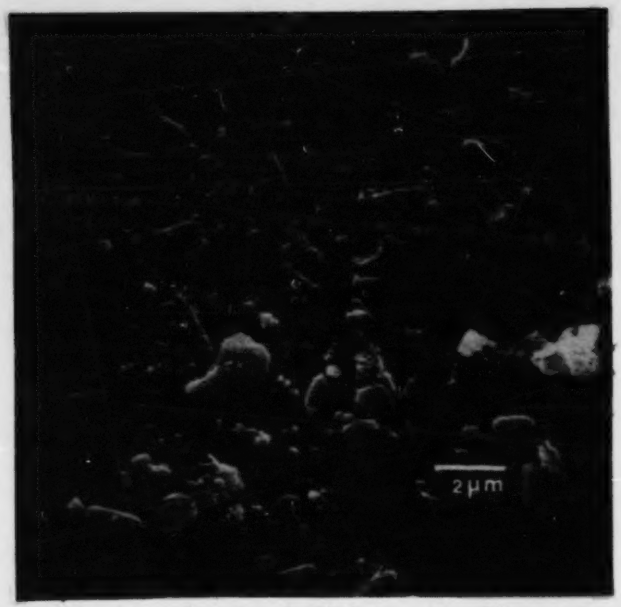

Figure C-8. Phase II: 50 ml of 24-hour standing water (5,250 X).

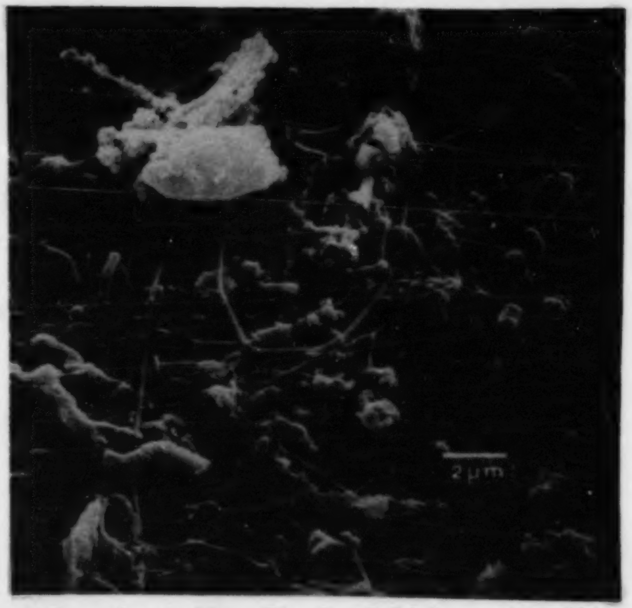

Figure C-9. Phase II: 50 ml of 24-hour standing water.
Note filaments and particulate debris (4,400 X).

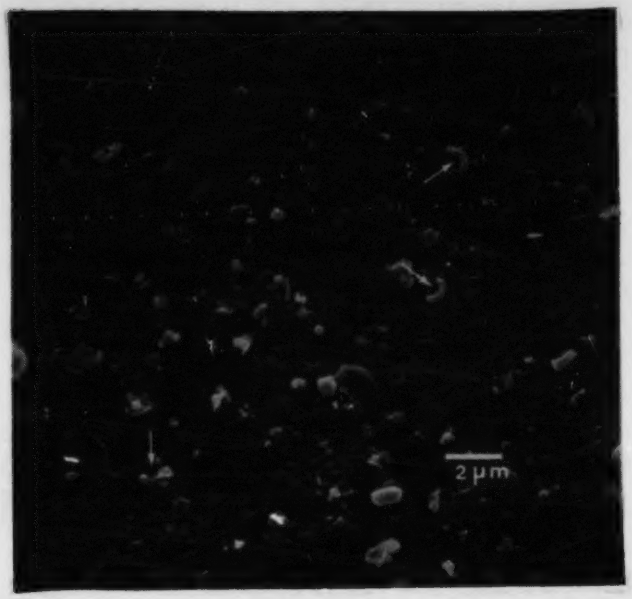

Figure C-10. Phase II: 50 ml of running water. Note vibrio-shaped bacteria (arrows) (4,330 X).

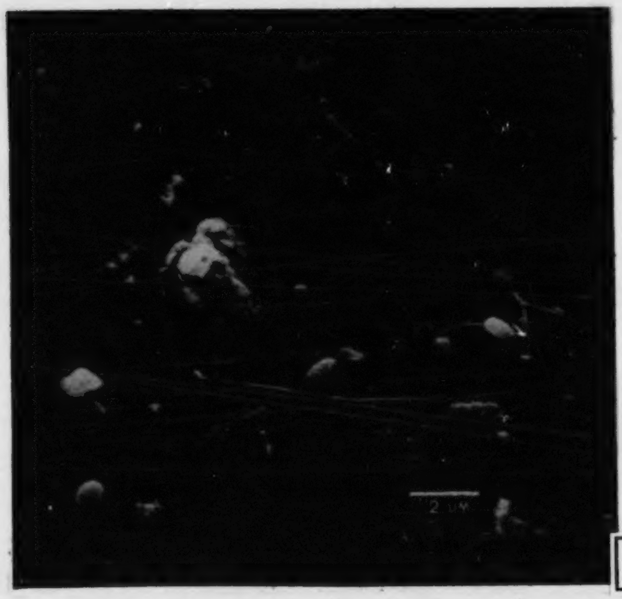

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Figure C-11. Phase II: 50 ml of running water. Note branching filaments (5,100 X).

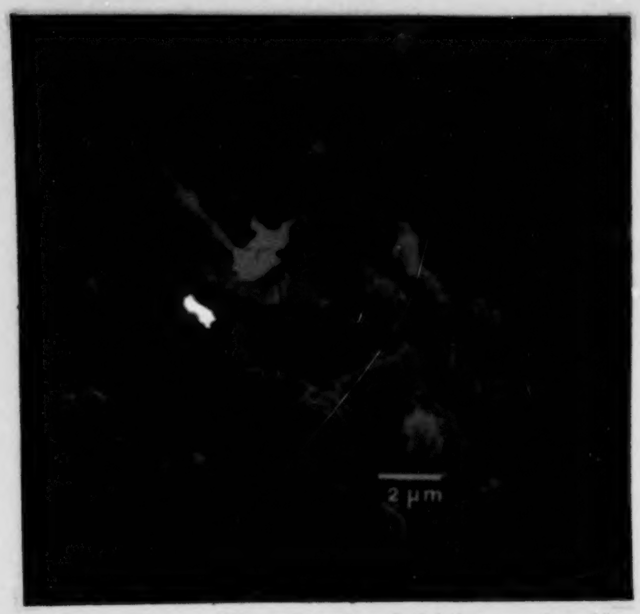

Figure C-12. Phase II: 20 ml of 24-hour standing water. Note amount of debris with embedded microbes (4,600 X).

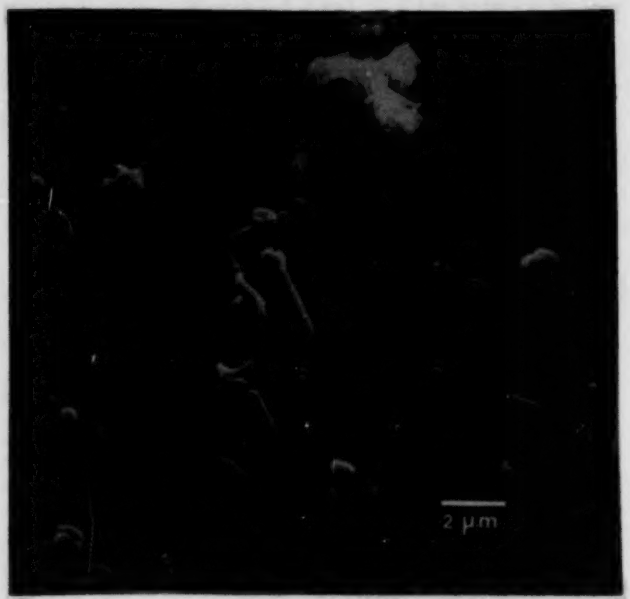

Figure C-13. Phase II: 50 ml of running water. Compare here the lack of particulate matter to that present in Plate 12 in only 20 ml of standing water. Both samples were collected at the same sampling time (4,400 X).

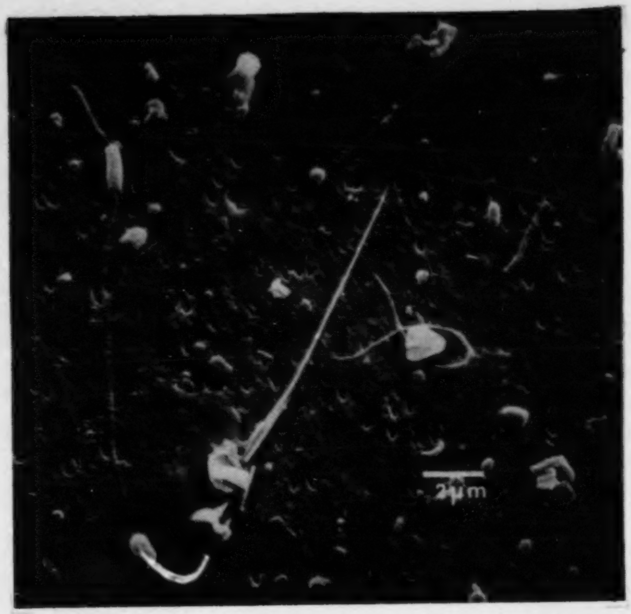

Figure C-14. Phase III: 50 ml of control water (4,350 X).

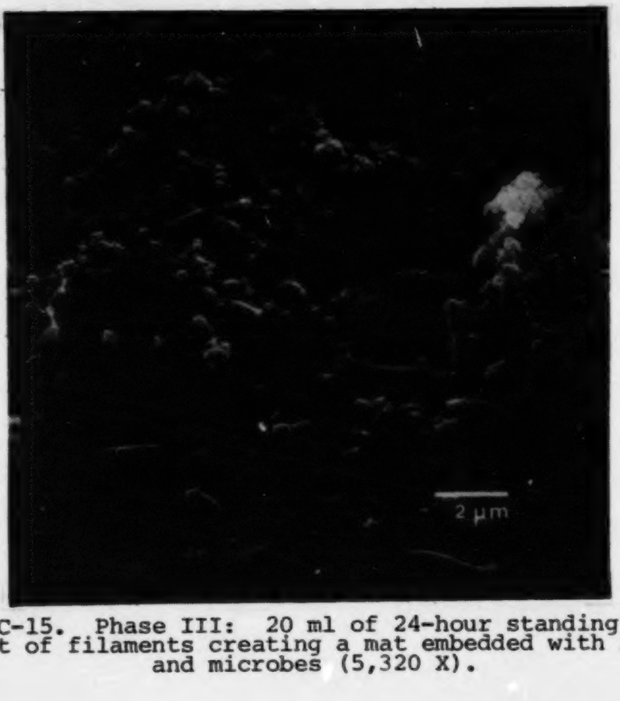

Figure C-15. Phase III: 20 ml of 24-hour standing water. Note large amount of filaments creating a mat embedded with inorganic debris and microbes (5,320 X).

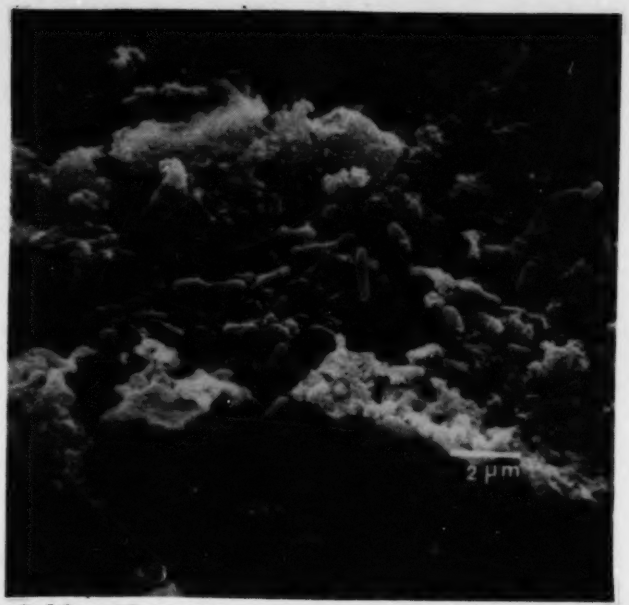

Figure C-16. Phase IV: 20 ml of 24-hour standing water. Note large numbers of coccoid and rod-shaped bacteria (4,900 X).

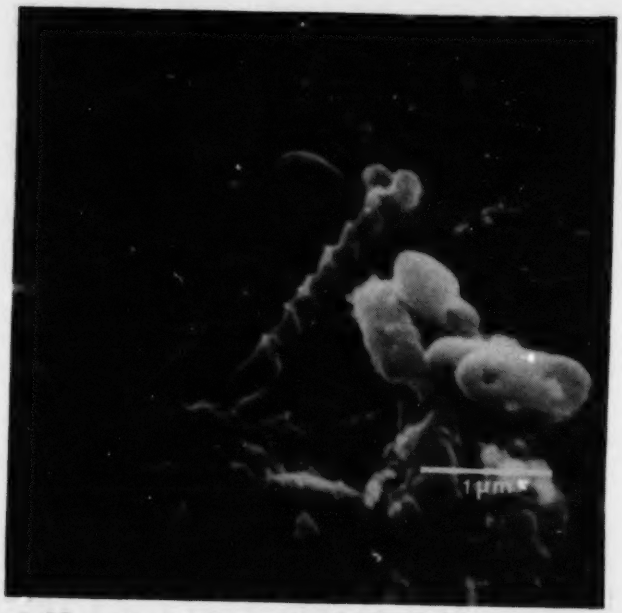

Figure C-17. Phase IV: 20 ml of 24-hour standing water. Note heavy blanket of filamentous bacteria (19,500 X).

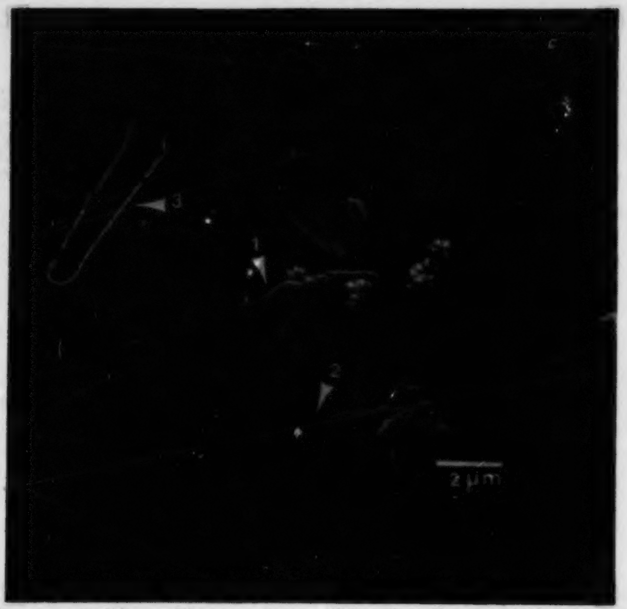

Figure C-18. Phase IV: 50 ml of running water. Note the presence of 1) a stalked bacterium, 2) rod-shaped bacteria, and 3) a broken diatom (4,910 X).

Figure C-19. Phase IV: 50 ml of control water. Noticeable absence of debris and bacteria as compared to running water, and especially to standing water (4,950 X).

Figure C-20. Phase IV: 20 ml of 24-hour standing water. Note thick layer of inorganic matter (5,450 X).

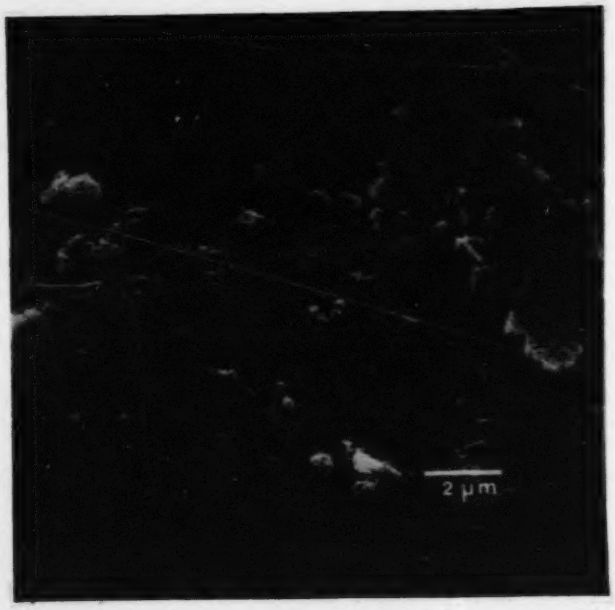

Figure C-21. Phase IV: 20 ml of 24-hour standing water. Note bacteria embedded in inorganic debris (5,530 X).

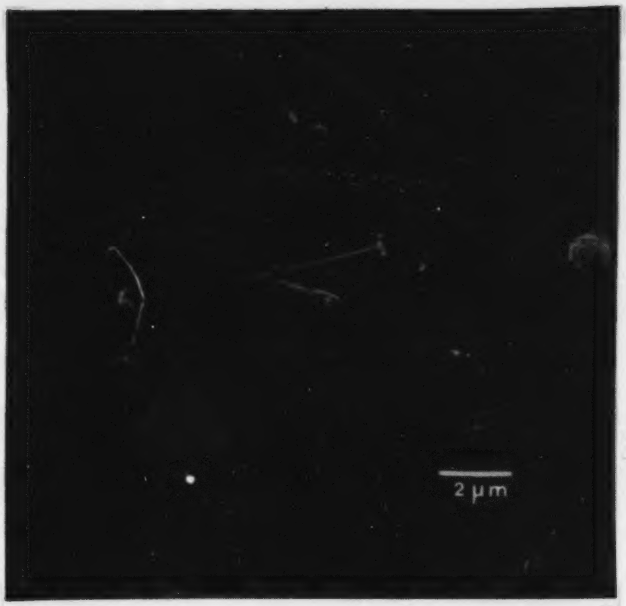

Figure C-22. Phase IV: 50 ml of running water. Note visible lack of inorganic precipitates and bacteria compared to standing water samples (5,400 X).

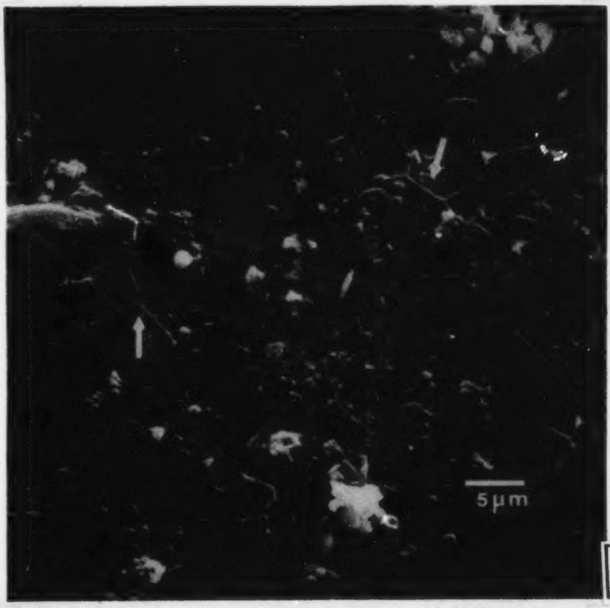

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Figure C-23. Running water sample displaying branched microorganisms (arrows), tentatively identified as Actinomycetes (1,770 X).

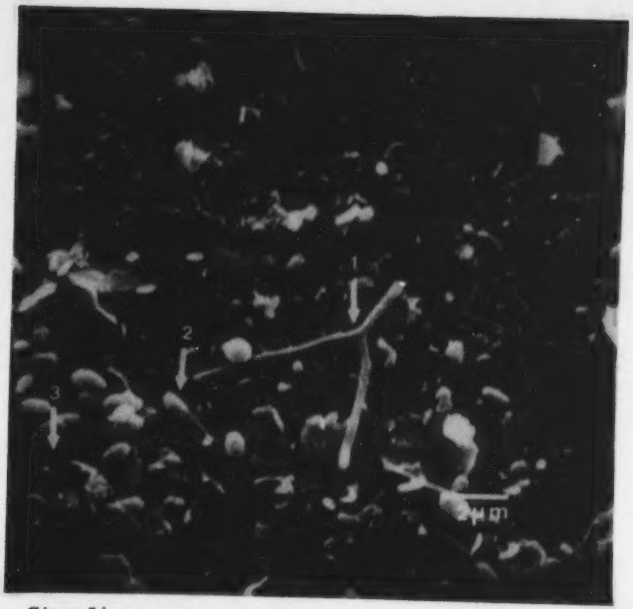

Figure C-24. Standing water sample displaying 1) a branched microbe, 2) a stalked bacterium, and 3) a possible member of the Cyanobacteria (5,150 X).

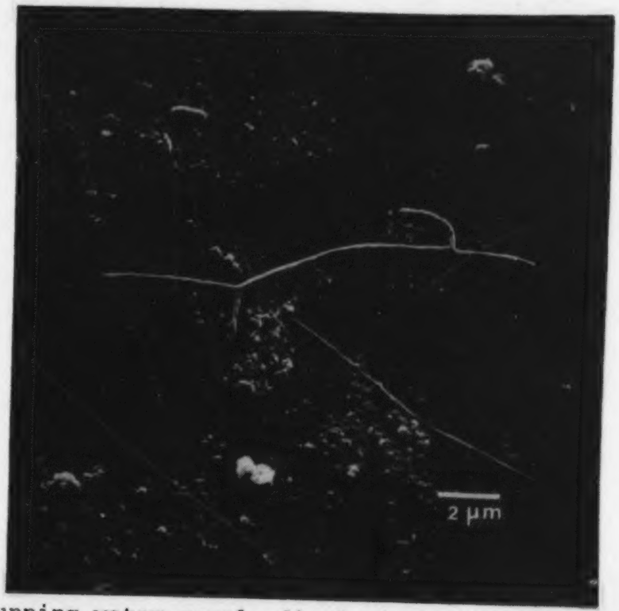

Figure C-25. Running water sample displaying a branched microorganism (4,500 X).

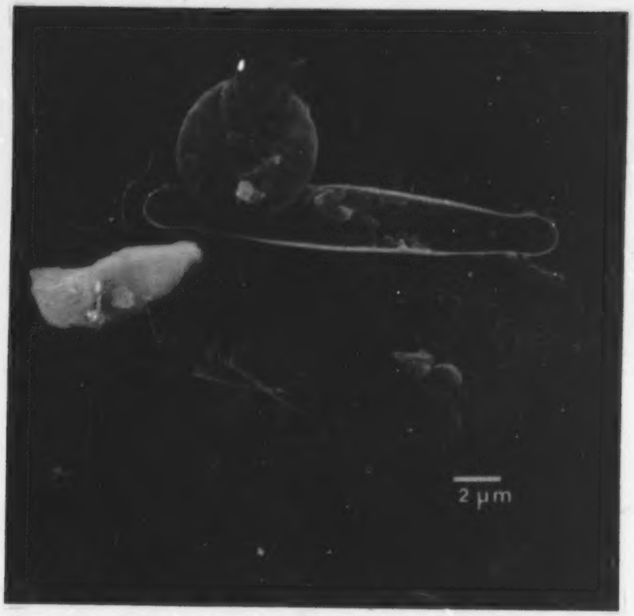

Figure C-26. Three species of diatoms with attached bacteria (3,560 X).

Figure C-27. Diatom with algae, organic and inorganic debris, and bacteria (4,430 X).

Figure C-28. Suspended particulate matter with attached microorganisms heavily coated with ferric deposits (9,450 X).

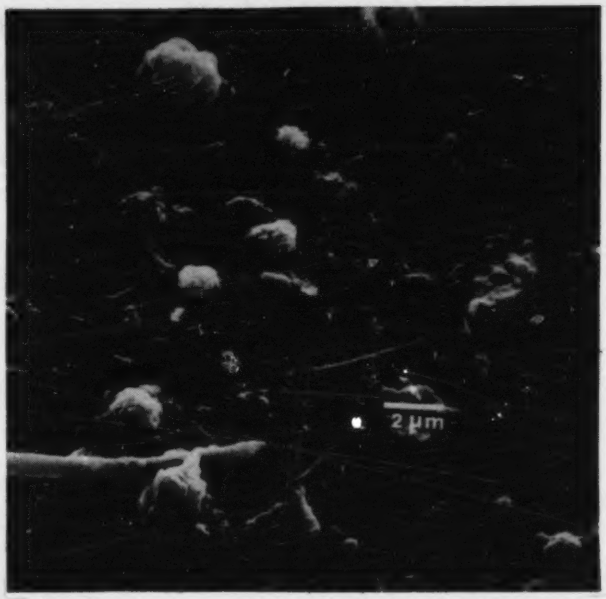

Figure C-29. Muccoid-appearing material with embedded and attached bacteria and particulate debris (4,450 X).

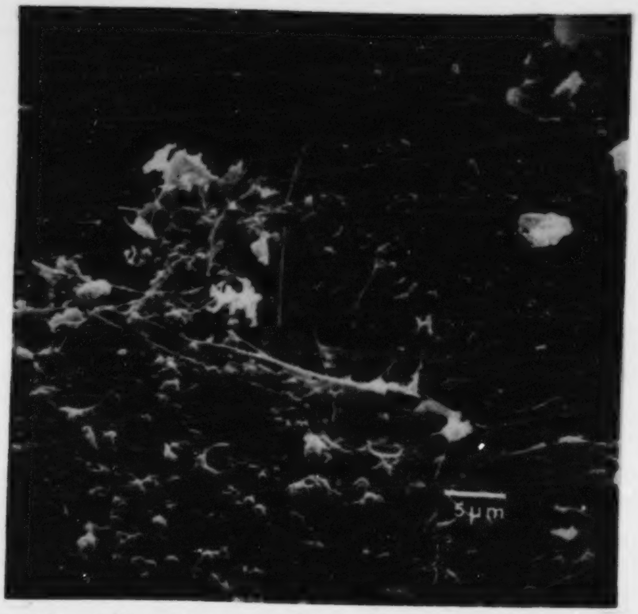

Figure C-30. Filamentous and stalked bacteria adhering to extracellular slime deposits (1,780 X).

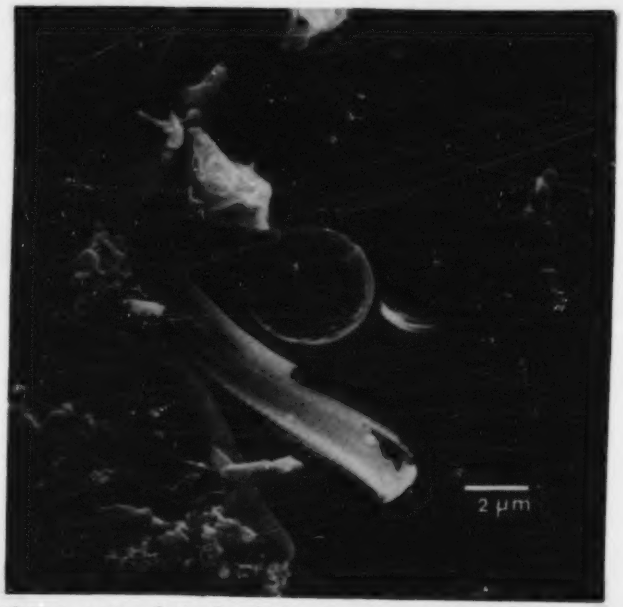

Figure C-31. Diatoms associated with algae and suspended particulate matter (4,910 X).

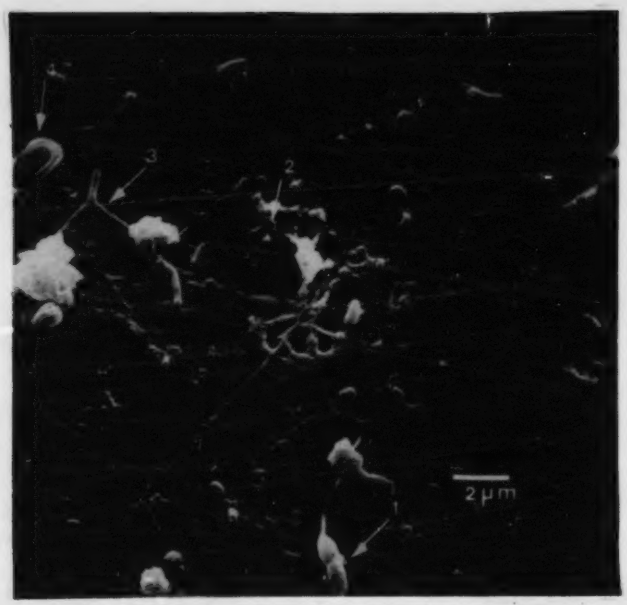

Figure C-32. A budding Hyphomicrobium sp. 1), seen with 2) a possible Cyanobacteria sp., 3) an Actinomycete, and 4) a spherical diatom (4,290).

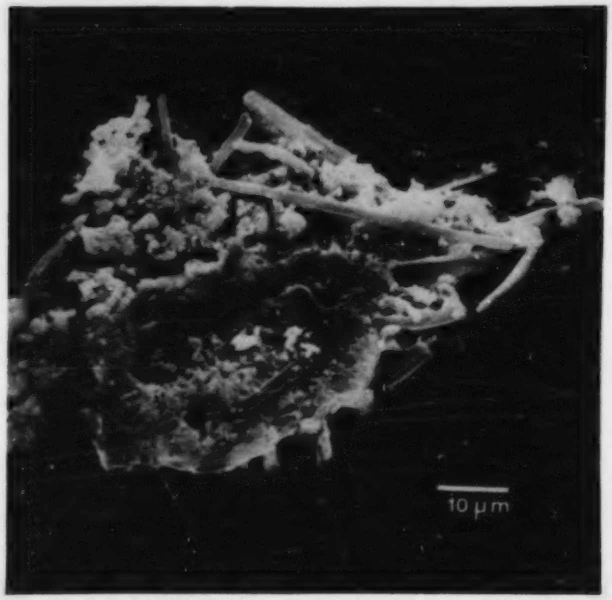

Figure C-33. Diatoms embedded in organic and inorganic material (1,030 X).

Figure C-34. Diatom sp. (arrow) with suspended particulate matter (4,640 X).

Figure C-35. A tentatively identified Caulobacter sp. isolated from selective culture medium. Arrows pointing to white crossbands (9,600 X).

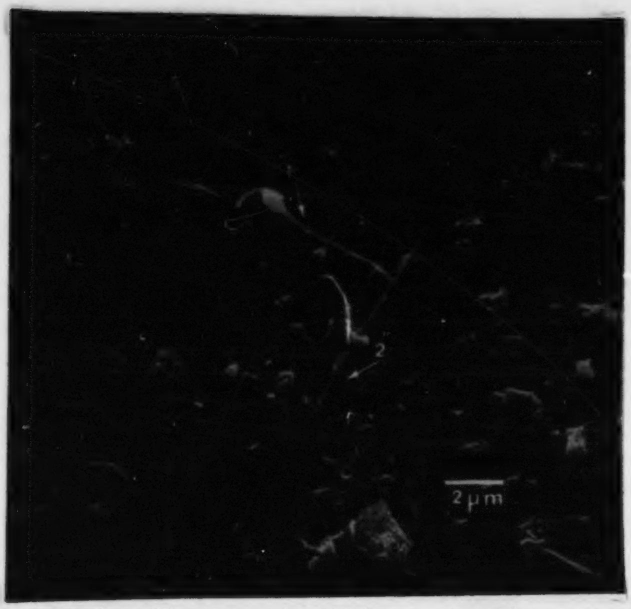

Figure C-36. A Hyphomicrobium sp. in 24-hour standing water (1). Another stalked bacterium, possibly a Caulobacter sp., is also viewed (2) (4,310).

Figure C-37. A Caulobacter sp. or Hyphomicrobium sp. observed in a control sample, Phase II (4,410 X).

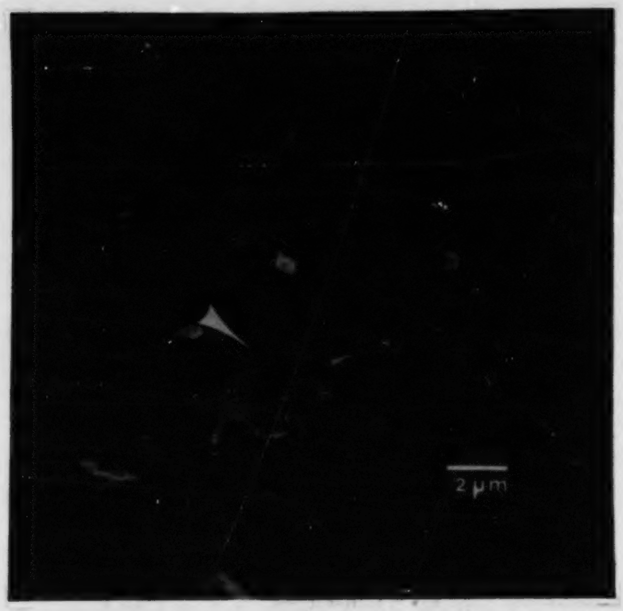

Figure C-38. A stalked bacterium, possibly a Hyphomicrobium sp., alongside a broken diatom (arrow) (4,700 X).

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Figure C-39. A Caulobacter sp. or Hyphomicrobium sp. found entwined with suspended particulate matter in 24-hour standing water (4,670 X).

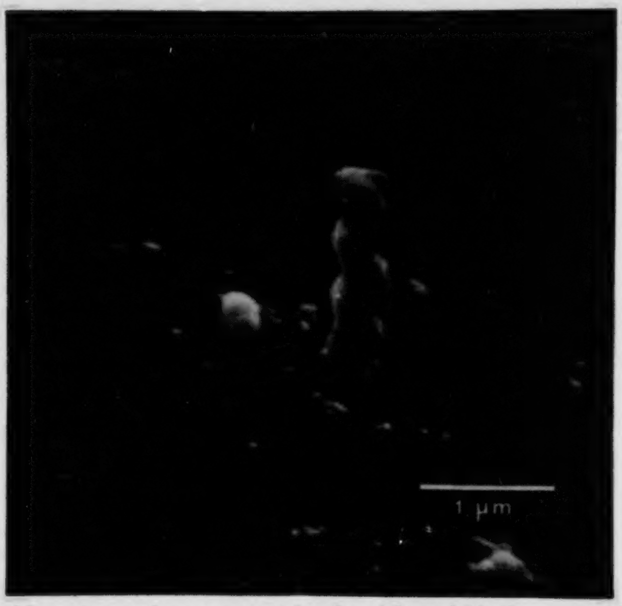

Figure C-40. Organism tentatively identified as Gallionella. Note depression in suspended particulate matter at top of stalk - most probably due to the missing cell (20,500 X).

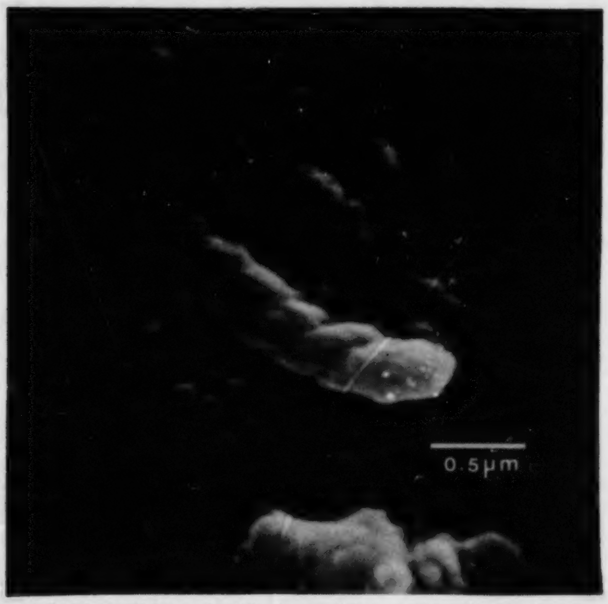

Figure C-41. A Gallionella stalk heavily coated with ferric hydroxide; obtained from a standing water sample (27,200 X).

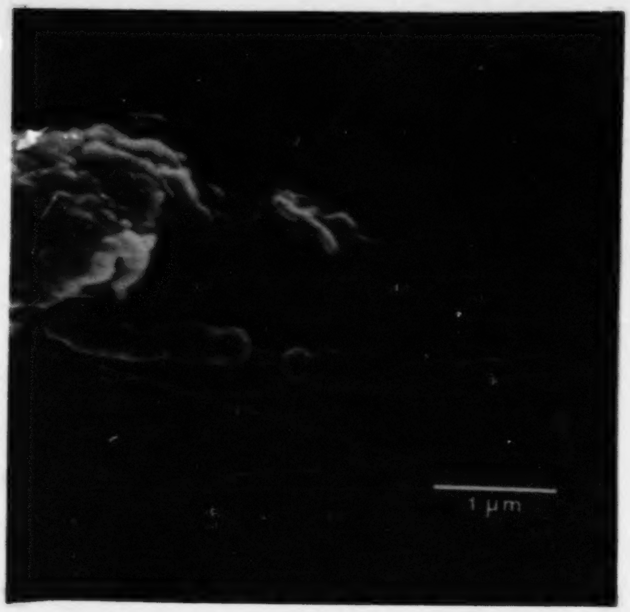

Figure C-42. Broken Gallionella stalk heavily coated with ferric hydroxide (18,700 X).

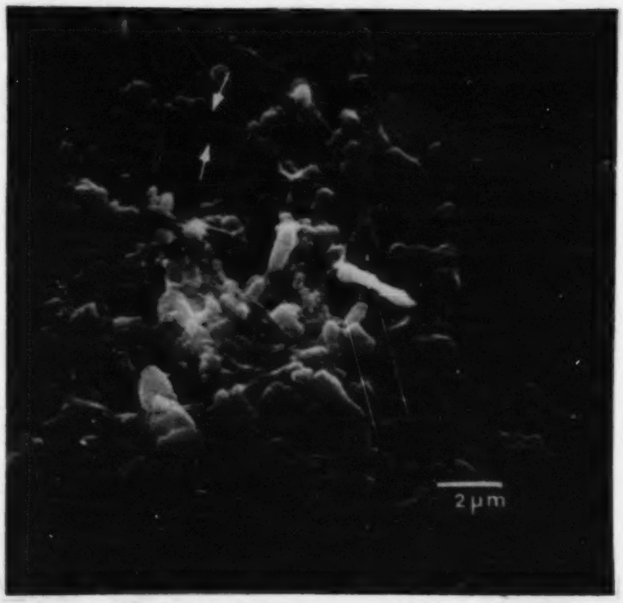

Figure C-43. View of Gallionella stalk (arrows) associated with other bacteria and particulate debris (5,130 X).

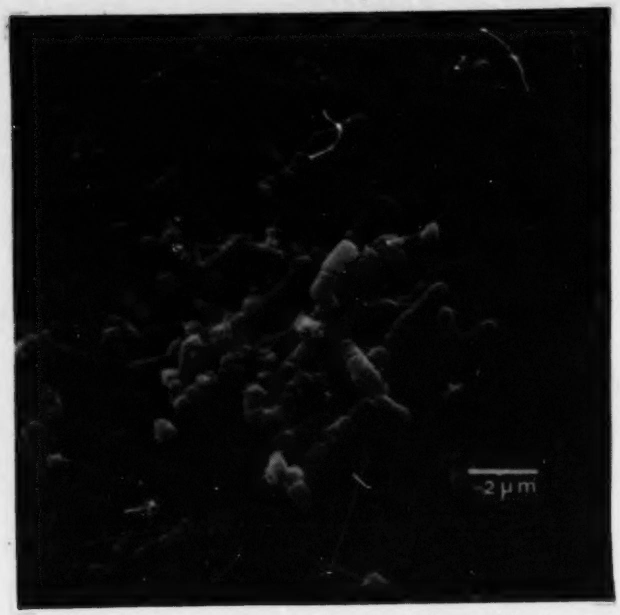

Figure C-44. Microorganisms tentatively identified as members of the *Sphaerotilus*-*Leptothrix* group (5,300 X).

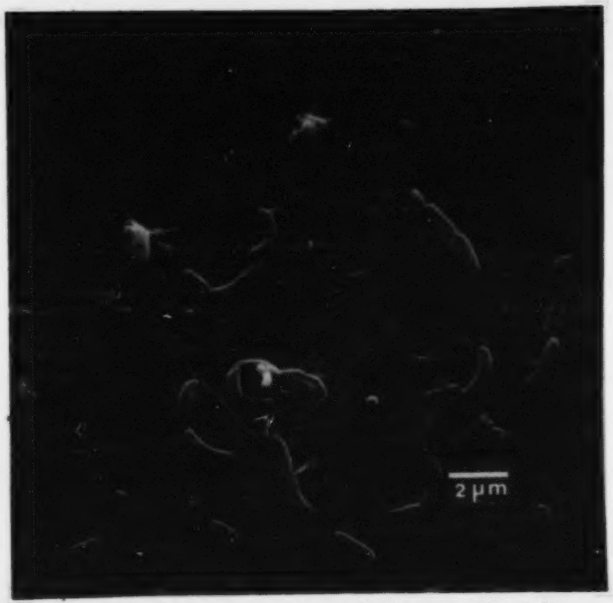

Figure C-45. *Desulfovibrio* isolated from selective culture media (4,840 X).

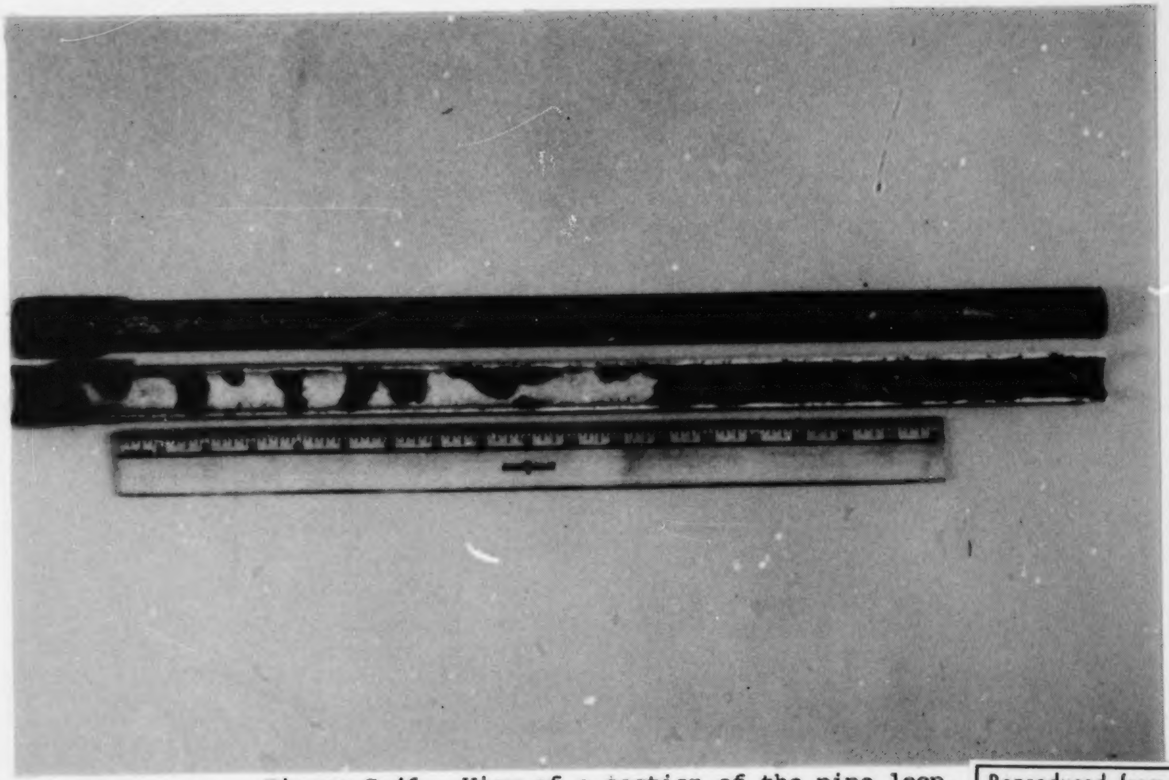

Figure C-46. View of a section of the pipe loop. Note the amount and size of the tubercles.

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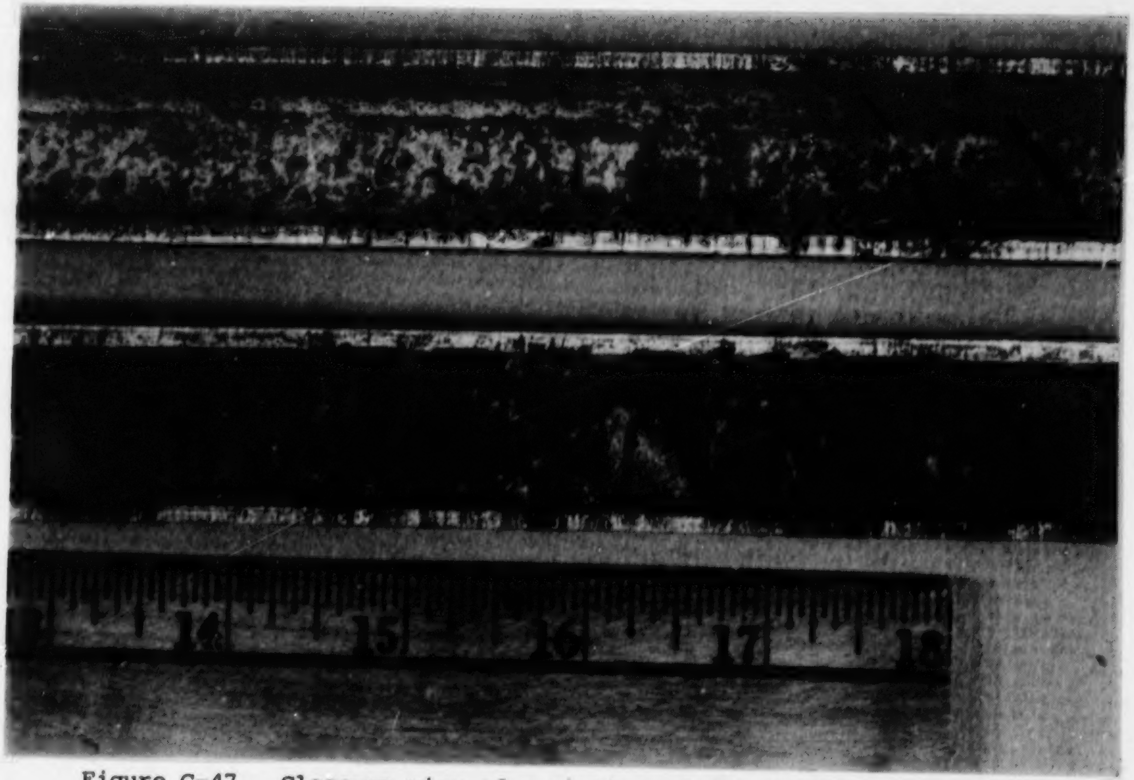

Figure C-47. Close-up view of a section of the pipe in Figure C-46. Some of the tubercles were up to 1 cm in height, while others left a pit up to 1.0 mm in depth.

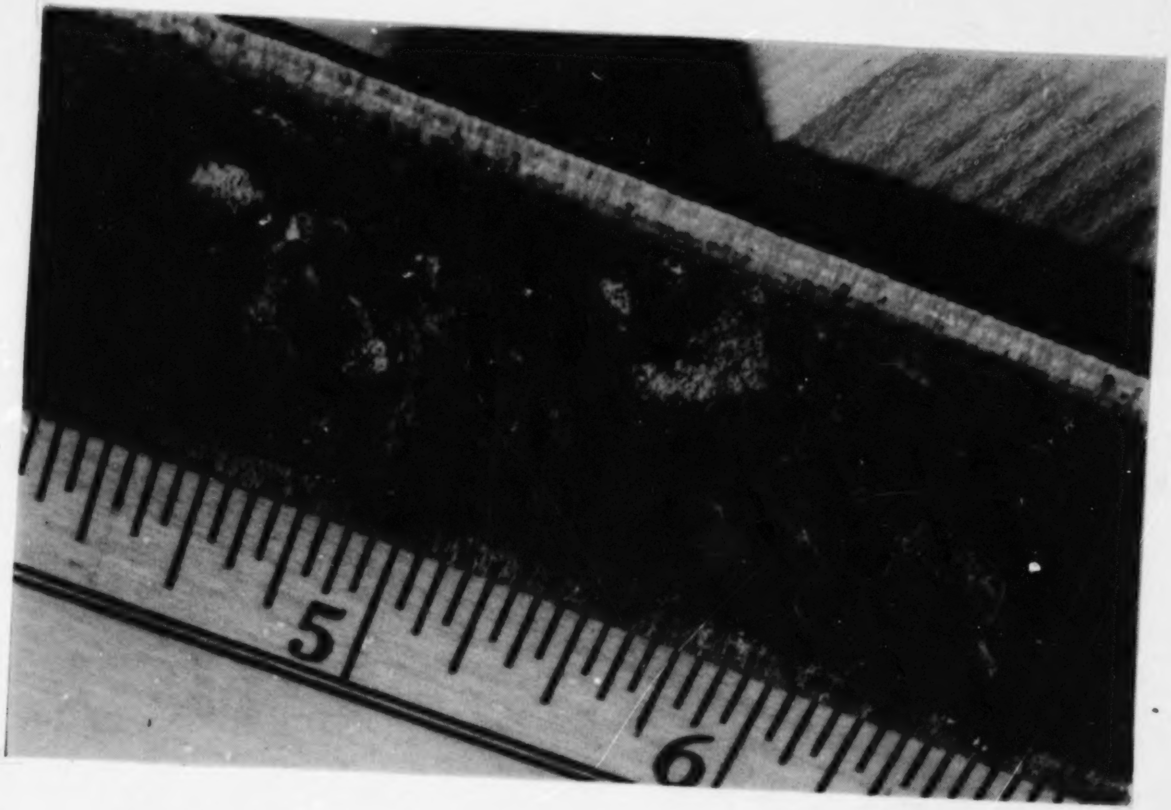

Figure C-48. Close-up view of several tubercles in Figure C-47.

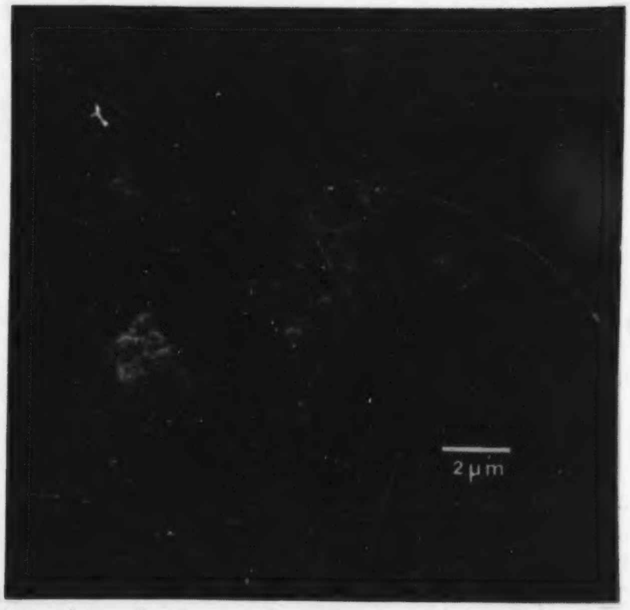

Figure C-49. Scanning electron micrograph of pipe incrustation surface showing superficial layer of flocculent-type precipitate (5,100 X).

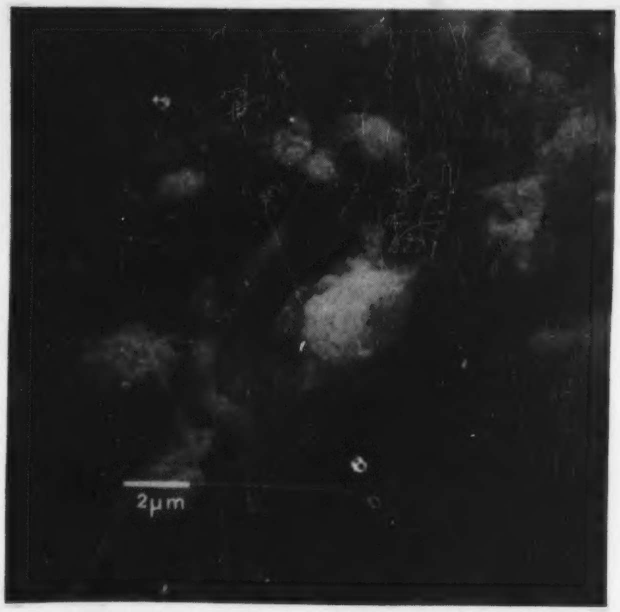

Figure C-50. View of heavily coated spiral-shaped bacteria attached to the mineral layer. Note the actinomycete-like filaments also adhering to the mineral layer (4,850 X).

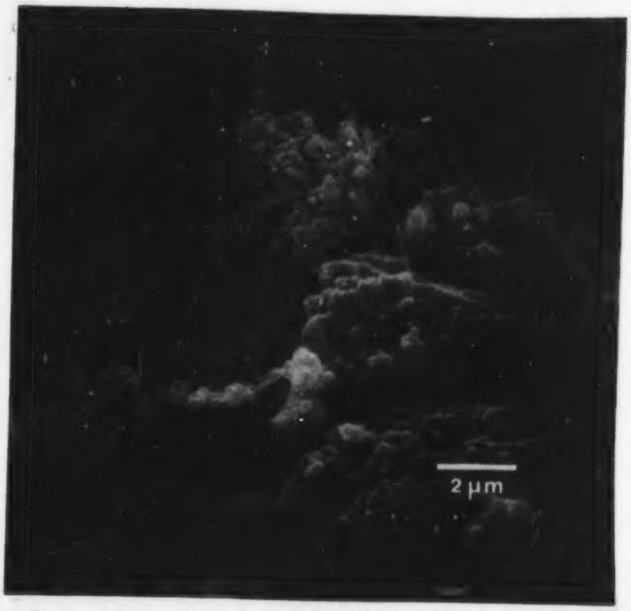

Figure C-51. Surface of rust-colored nodule showing flocculent precipitate (5,930 X).

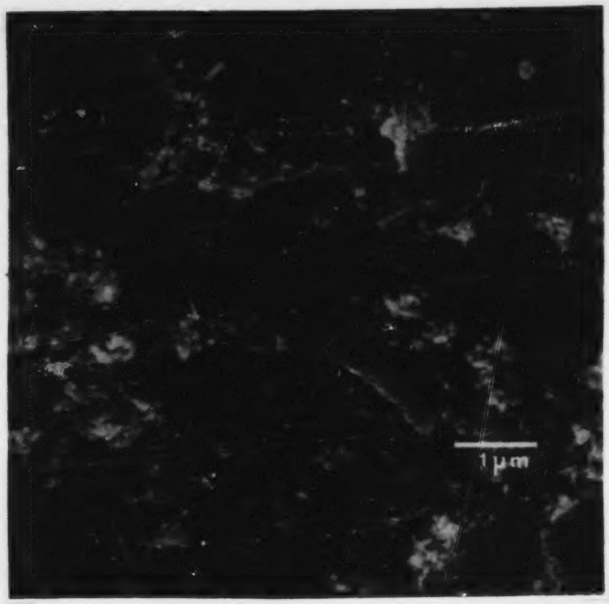

Figure C-52. Tubercular surface showing a thick mineral layer and attached bacterium (12,000 X).

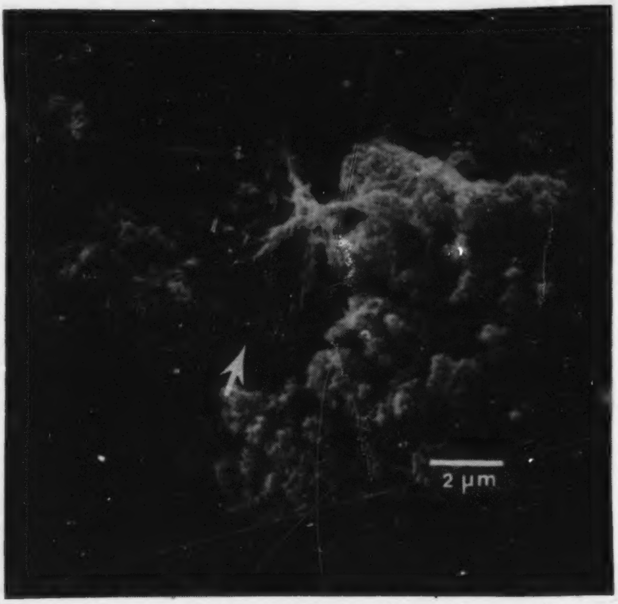

Figure C-53. Region of microcolonization by actinomycete-like filamentous microbes (arrow), (5,380 X).

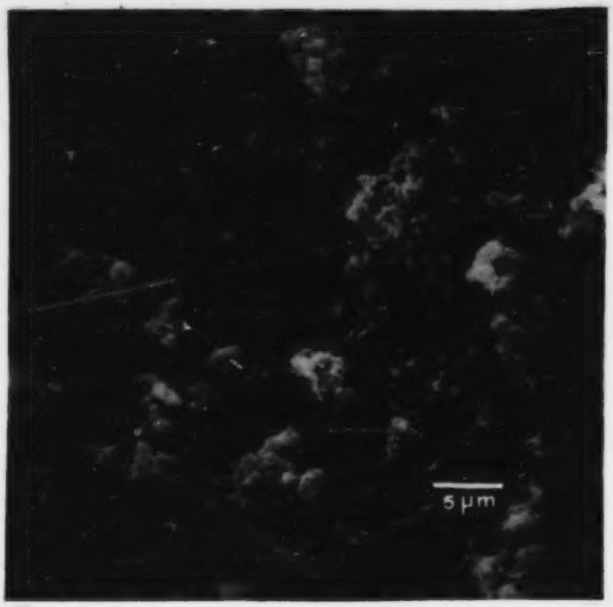

Figure C-54. Actinomycete-like filaments adhering to mineral layer (2,060 X).

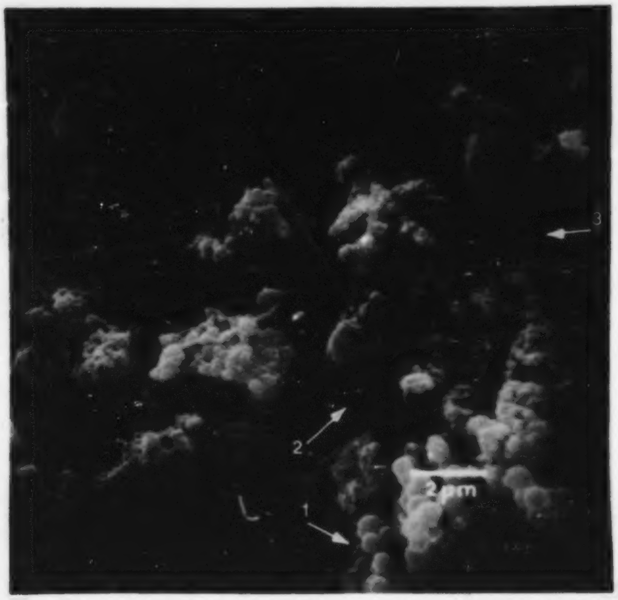

Figure C-55. Microcolony of coccoid-shaped bacteria (1). Note the smooth surface appearance of the individual cells. The mineral layer also shows entrapment of other bacteria, (2) and (3), (5,700 X).

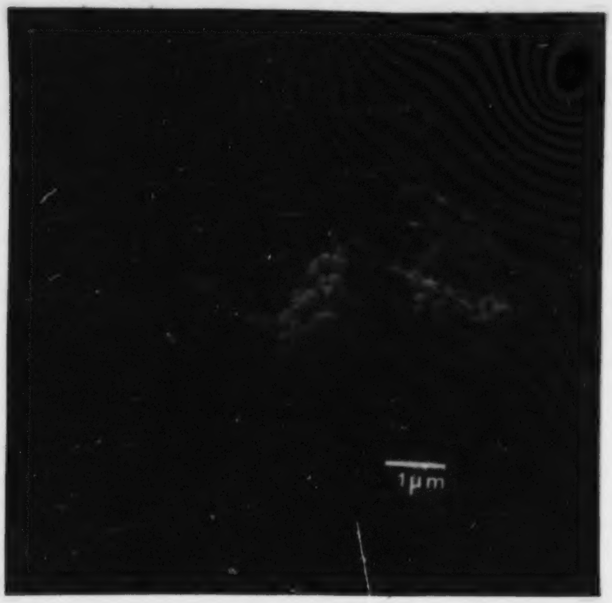

Figure C-56. Region of dense microcolonization by filamentous antinomycete-like microbes (9,100 X).

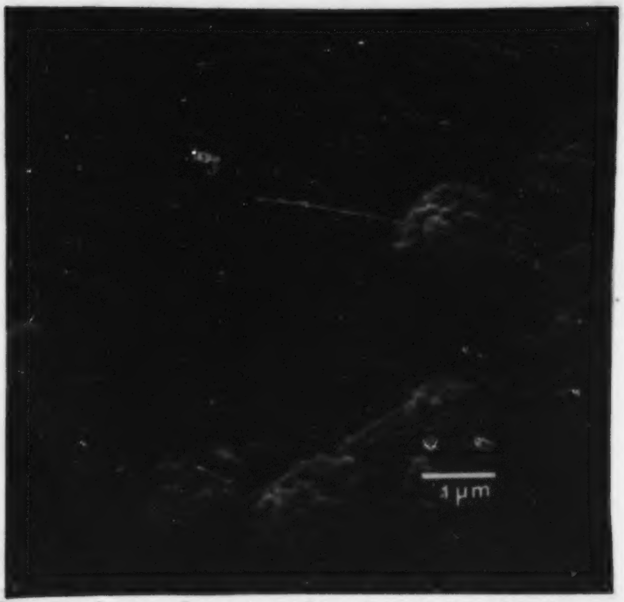

Figure C-57. Surface of rust-colored nodule showing trapped diatom and filaments (11,150 X).

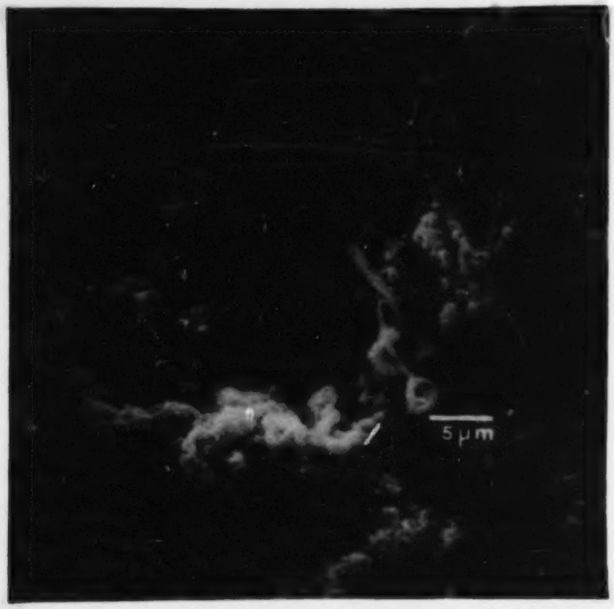

Figure C-58. Mineral layer showing attachment of filamentous microbes and other bacteria (1,920 X).

Figure C-59. Region of microcolonization of filamentous microbes (2,140 X).

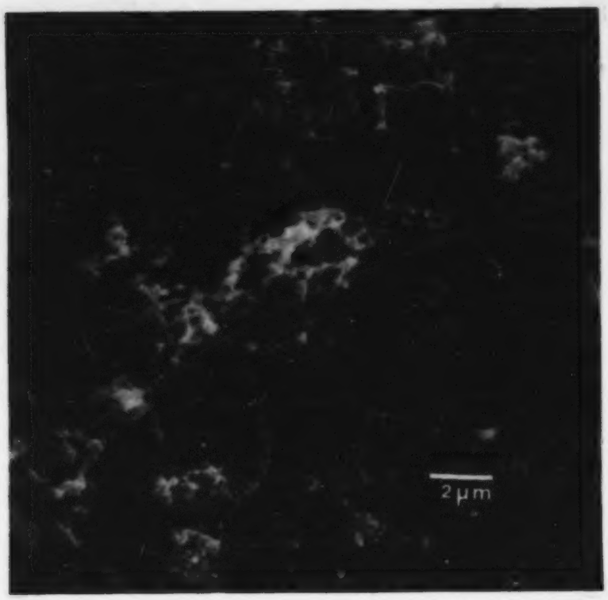

Figure C-60. Filamentous bacteria coated with flocculent precipitate and firmly anchored to mineral layer (4,700 X).

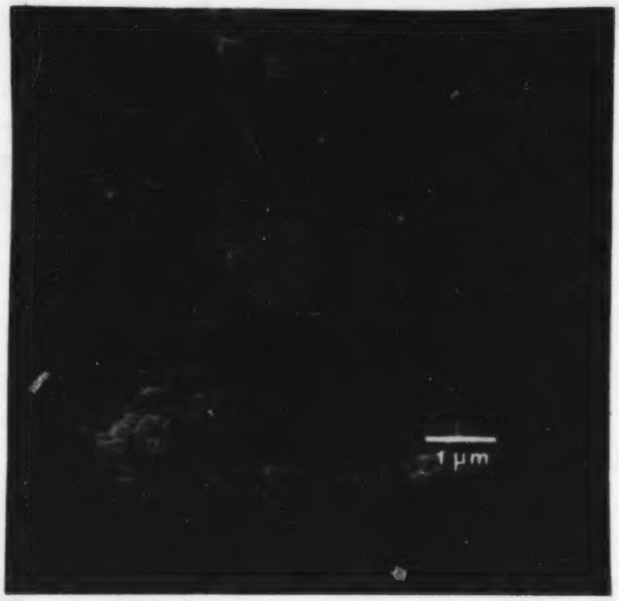

Figure C-61. Ferric hydroxide nodules attached to a tentatively identified heavily coated Gallionella stalk (10,200 X).

Figure C-62. Small spherical nodules of ferric hydroxide adhering to stalked bacteria (11,250 X).

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Figure C-63. Rust-colored nodule treated with 1% oxalic acid for 10 minutes. Note granular appearance of ferric precipitate and attachment of bacteria (arrows), (4,750 X).

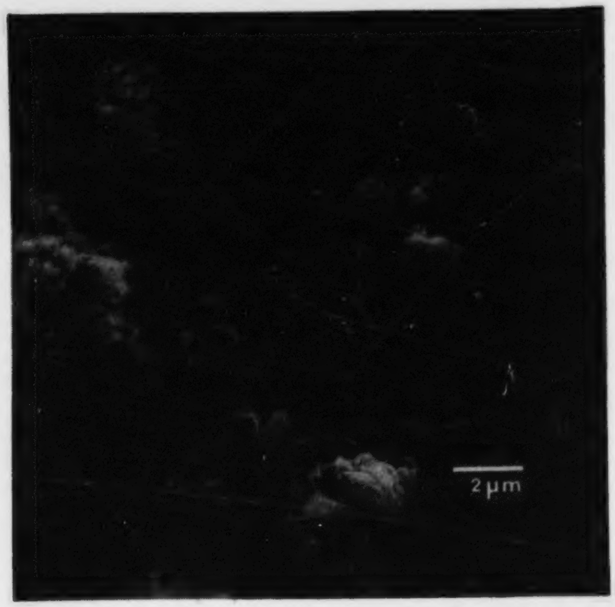

Figure C-64. Tubercle treated with 1% oxalic acid for 15 minutes (5,100 X).

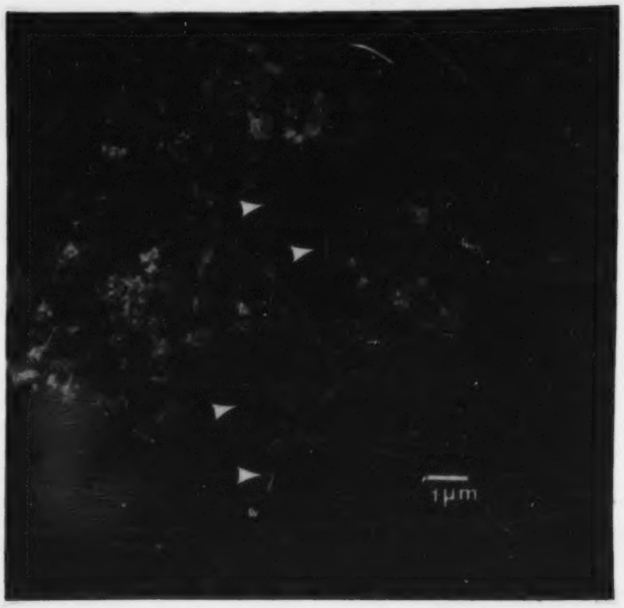

Figure C-65. Pipe incrustation treated with 5% oxalic acid for 10 minutes. Note attached bacteria (6,050 X).

Figure C-66. Tubercle treated with 5% oxalic acid for 15 minutes. Note granular appearance of the mineral layer. Filamentous microbes and rod-shaped bacteria were becoming visible (arrows), (2,360 X).

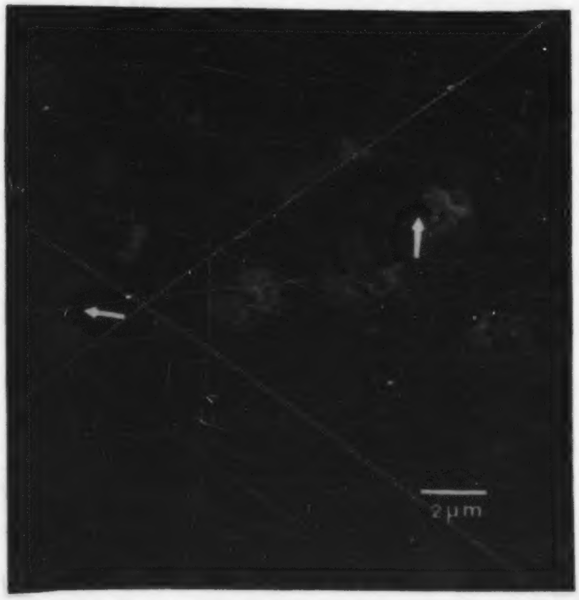

Figure C-67. Tubercle treated with 5% oxalic acid for 15 minutes. Note appearance of vibrio-shaped bacteria (arrows) and filaments (4,500 X).

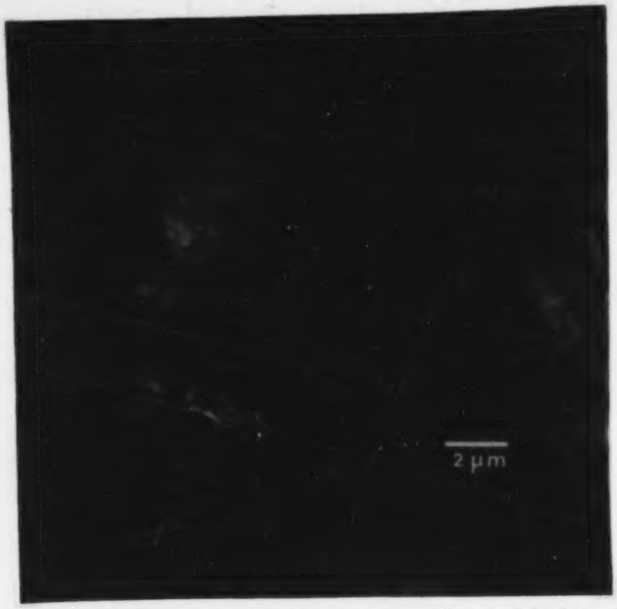

Figure C-68. Tubercle treated with 5% oxalic acid for 20 minutes. As the ferric precipitate was dissolved, embedded filaments and bacteria became visible (4,670 X).

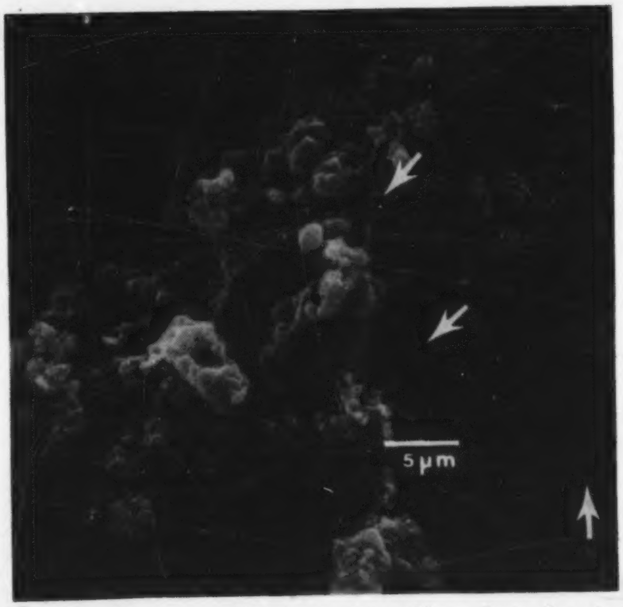

Figure C-69. Extracellular fibrils extruding from bacteria (arrows) (2,220 X).

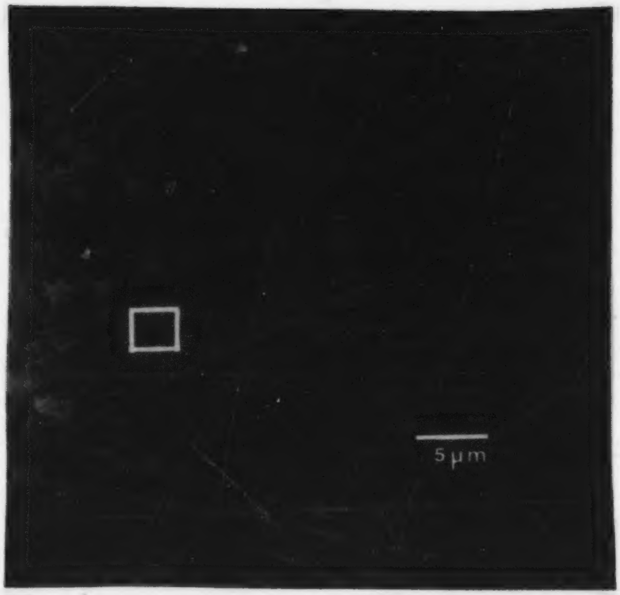

Figure C-70. Crevice pipe incrustation showing microcolonization of filamentous bacteria (2,240 X).

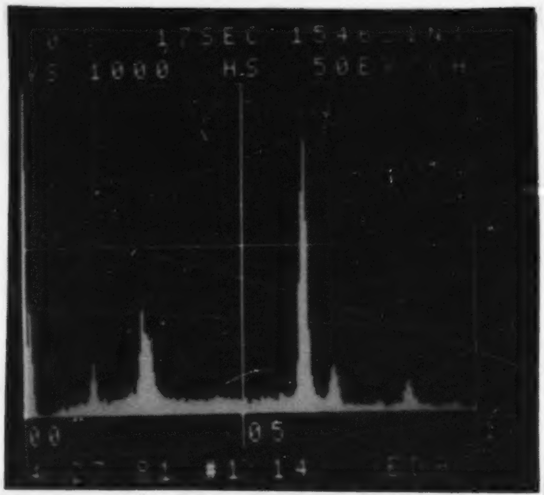

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Figure C-71. X-ray energy-dispersive microanalysis of tubercle in Figure C-70. Box indicates area where electron beam was focused. Relevant peaks were identified as follows: K_{α} line of aluminum (1.49 keV); K_{α} line of silicon (1.74 keV); K_{α} line of iron (6.40 keV); K_{α} line of iron (7.06 keV); and K_{α} line of zinc (8.64 keV). X-ray counts were integrated over a period of 17 seconds. (Note: The large peak seen at approximately 3.0 keV is exhibiting the Au/Pd with which the specimen was coated.)

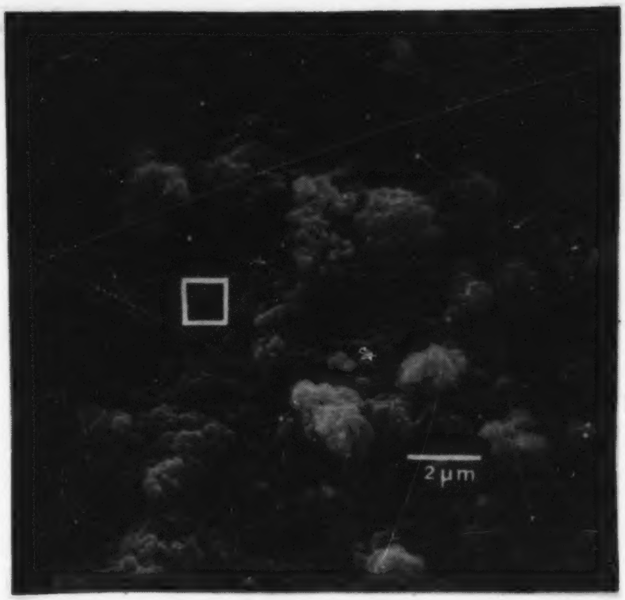

Figure C-72. Rust-colored nodule surface showing heavily deposited mineral layer (5,600 X).

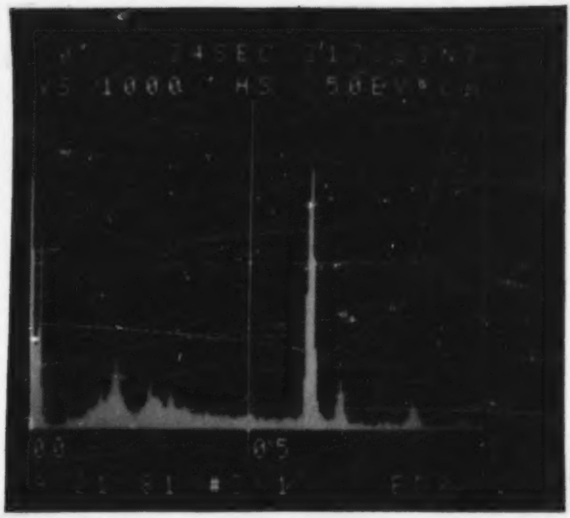

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Figure C-73. X-ray energy dispersive microanalysis of mineral layer in Figure C-72. Box indicates region where electron beam was focused. Relevant peaks were identified as follows: L_α line of zinc (1.001 keV); K_α line of silicon (1.74 keV); K_α line of phosphorous (2.01 keV); K_α line of calcium (3.69 keV); K_α line of manganese (5.89 keV); K_α line of iron (6.40 keV); K_β line of iron (7.06 keV); K_α line of zinc (8.64 keV); and K_β line of zinc (9.62 keV). X-ray counts were integrated over a period of 34 seconds.

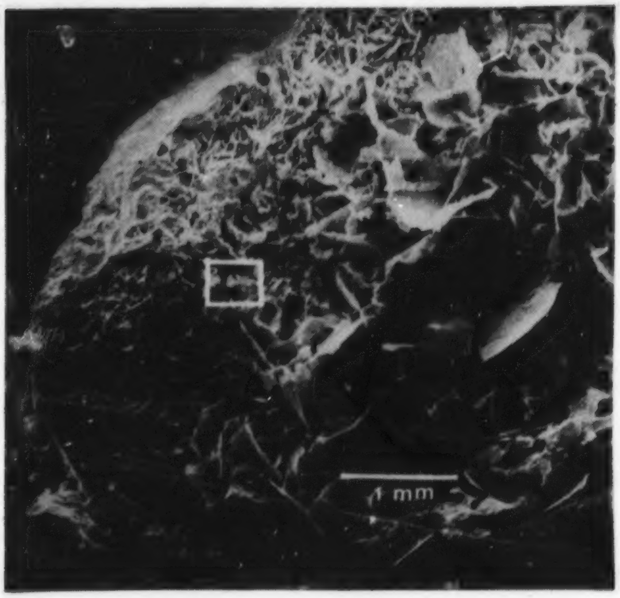

Figure C-74. View of tubercle interior showing large lamellar crystals of green rust (18.0 X).

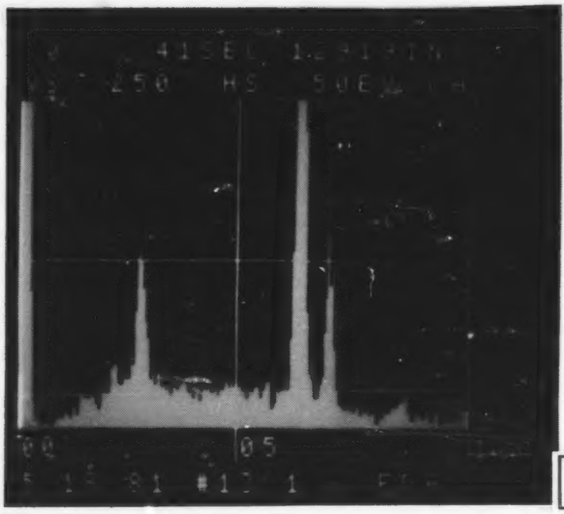

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Figure C-75. Expanded X-ray energy scan of tubercle interior in Figure C-74. Box indicates region where electron beam was focused. Relevant peaks were identified as follows: K_α line of aluminum (1.49 keV); K_α line of sulfur (2.31 keV); K_α line of iron (6.40 keV); K_β line of iron (7.06 keV); and K_α line of zinc (8.64 keV). X-ray counts were integrated over a period of 41 seconds.

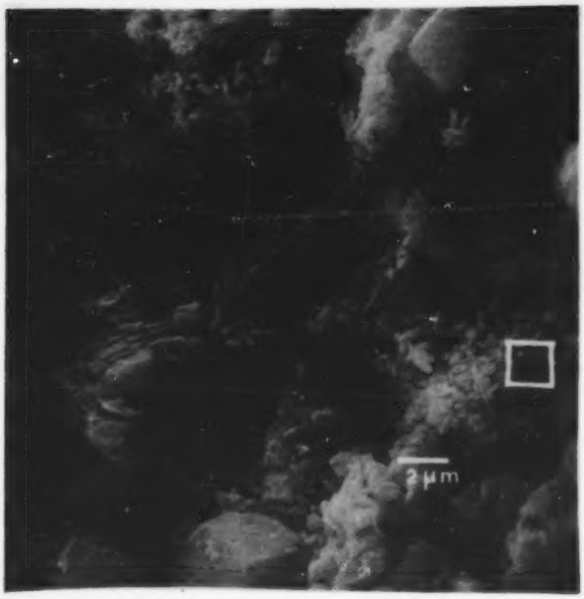

Figure C-76. Surface of tubercle after treatment with 5% oxalic acid for 10 minutes (3,700 X).

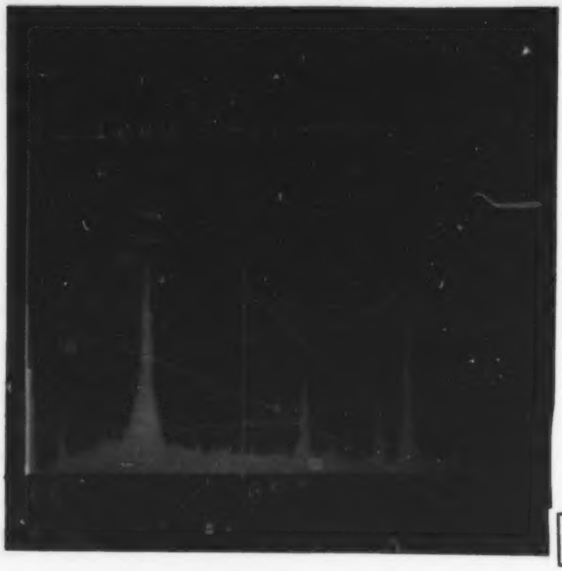

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Figure C-77. X-ray energy scan of area enclosed by box in Figure C-76. Relevant peaks were identified as follows: L_α line of zinc (1.001 keV); K_α line of aluminum (1.49 keV); K_α line of phosphorous (2.01 keV); K_α line of sulfur (2.31 keV); K_α line of chlorine (2.62 keV); K_α line of iron (6.40 keV); K_β line of iron (7.06 keV); K_α line of copper (8.04 keV); K_α line of zinc (8.64 keV); and K_β line of zinc (9.62 keV). X-ray counts were integrated over a period of 25 seconds.

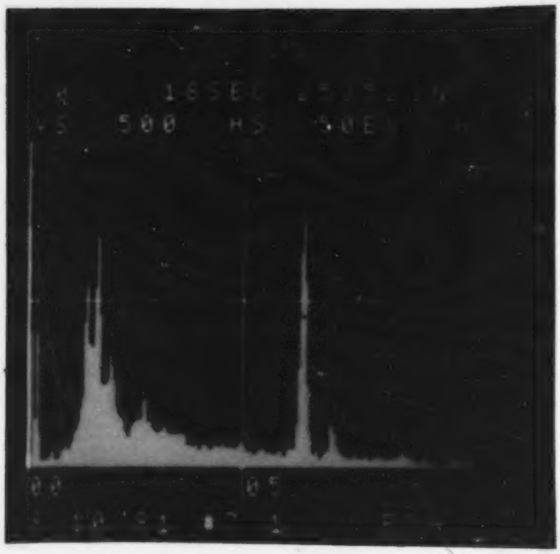

Figure C-79. X-ray energy scan of area enclosed by box in Figure C-78. Relevant peaks were identified as follows: L_α line of zinc (1.001 keV); K_α line of aluminum (1.49 keV); K_α line of silicon (1.74 keV); K_α line of phosphorous (2.01 keV); K_α line of manganese (5.89 keV); K_α line of iron (6.40 keV); and K_β line of iron (7.06 keV). X-ray counts were integrated over a period of 18 seconds.

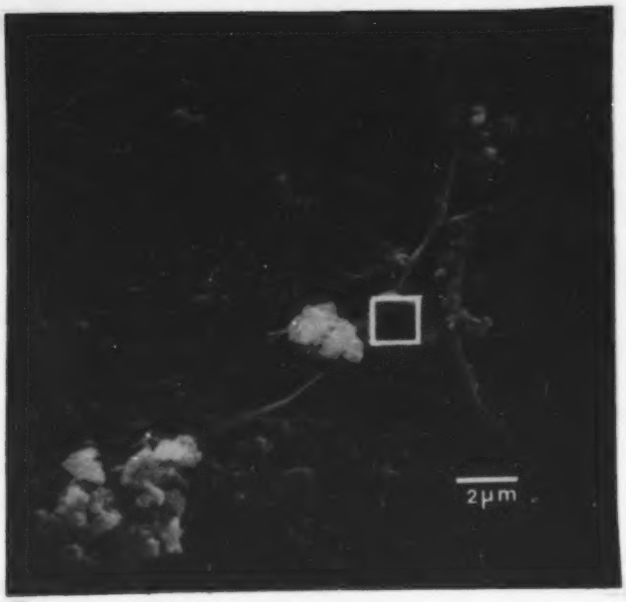

Figure C-78. Phase IV: 20 ml of 24-hour standing water (4,520 X).

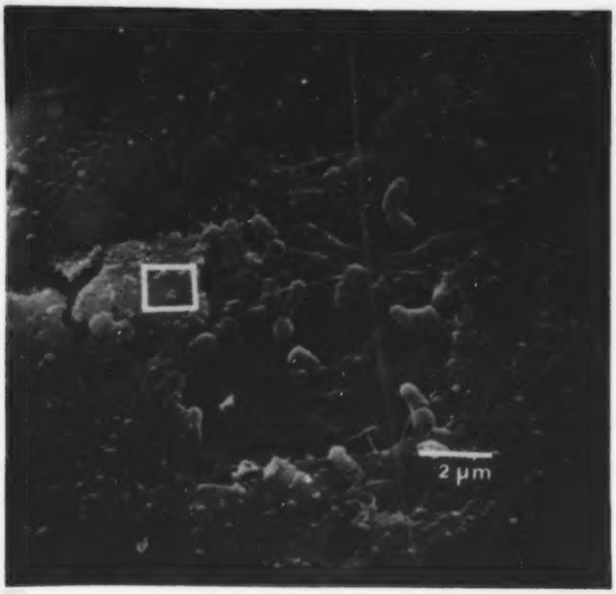

Figure C-80. Phase IV: 20 ml of 24-hour standing water (5,520 X).

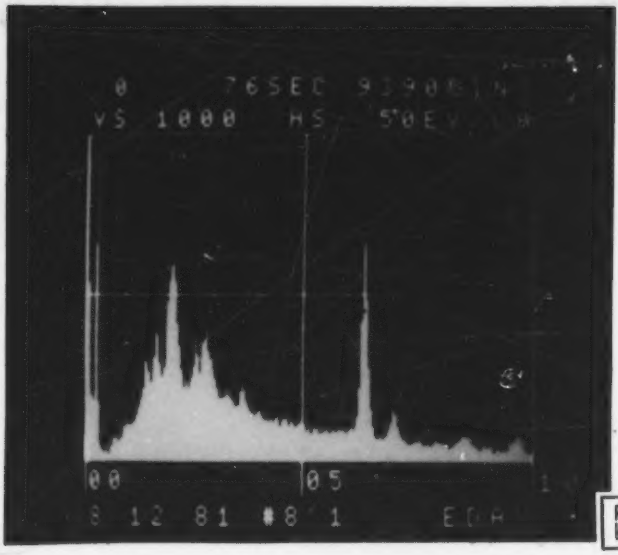

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Figure C-81. X-ray energy scan of area enclosed by box in Figure C-80. Relevant peaks were identified as follows: L line of zinc (1.001 keV); K_α line of aluminum (1.49 keV); K_α line of silicon (1.74 keV); K_α line of phosphorous (2.01 keV); K_α line of sulfur (2.31 keV); K_α line of chlorine (2.62 keV); K_α line of potassium (3.31 keV); K line of calcium (3.69 keV); K_α line of iron (6.40 keV); K_β line of iron (7.04 keV); K_α line of zinc (8.64 keV); and K_β line of zinc (9.62 keV). X-ray counts were integrated over a period of 76 seconds.

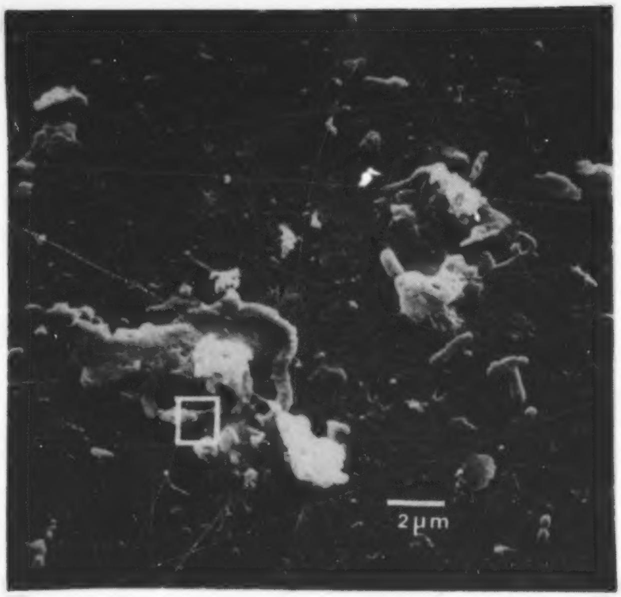

Figure C-82. Phase IV: 50 ml of running water (4,400 X).

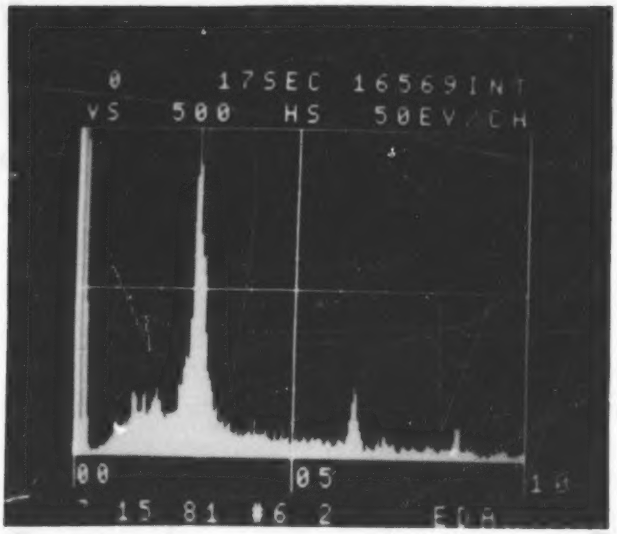

Figure C-83. X-ray energy scan of area enclosed by box in Figure C-82. Relevant peaks were identified as follows: K_α line of aluminum (1.49 keV); K_α line of silicon (1.74 keV); K_α line of phosphorous (2.01 keV); K_α line of chlorine (2.62 keV); K_α line of iron (6.40 keV); K_β line of iron (7.06 keV); and K_α line of zinc (8.64 keV). X-ray counts were integrated over a period of 17 seconds.

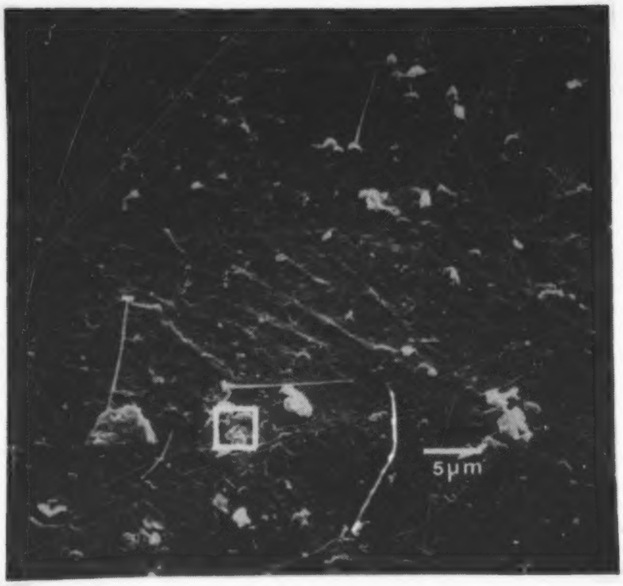

Figure C-84. Phase IV: 50 ml of control water (1,700 X).

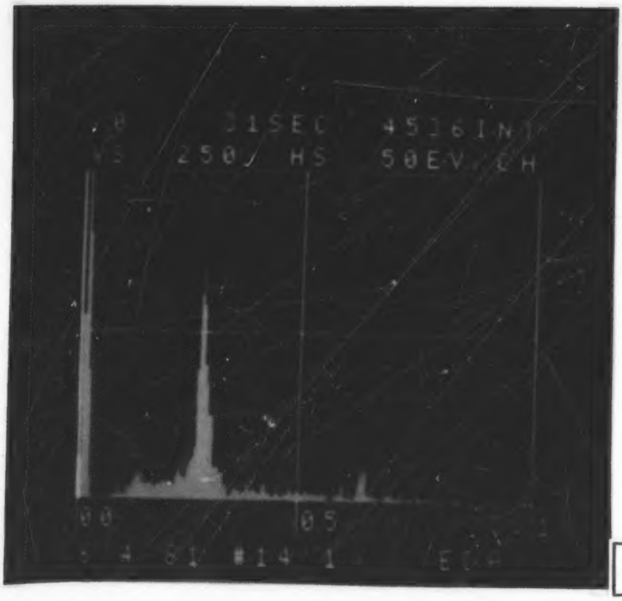

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Figure C-85. X-ray energy scan of area enclosed by box in Figure C-84. Relevant peaks were identified as follows: K_α line of aluminum (1.49 keV); K_α line of silicon (1.74 keV); and K_α line of iron (6.40 keV). X-ray counts were integrated over a period of 31 seconds.

APPENDIX D

RUNNING WATER METALS RESULTS FROM THE ON-SITE TREATMENT TESTS

Treatment	Iron mg/L	Lead µg/L	Zinc mg/L	Cadmium µg/L	Copper mg/L
Control	0.40	1.0	0.33	0.10	0.070
"	0.80	1.5	0.56	0.10	0.085
"	0.37	1.5	0.27	0.05	0.19
"	0.68	1.5	0.43	<0.05	0.06
"	0.51	1.5	0.27	0.10	
"	0.22	1.0	0.24	0.10	0.065
"	0.13	2.5	0.30	0.10	0.12
"	0.24	1.5	0.26	0.15	0.06
"	0.51	4.0	0.35	0.15	0.09
"	0.32	1.5	0.33	0.10	0.17
"	0.19	1.0	0.22	0.10	0.09
"	0.26	1.5	0.30	0.10	0.295
Na_2CO_3	0.27	2.0	0.22	0.05	0.26
"	0.14	1.0	0.085	<0.05	0.15
"	0.21	1.0	0.17	0.10	0.28
"	0.11	1.5	0.12	0.10	
"	0.11	1.0	0.12	0.10	0.06
"	0.24	1.5	0.19	0.05	0.14
"	0.16	1.0	0.12	0.05	0.15
"	0.22	1.5	0.12	<0.05	0.15
"	0.10	1.0	0.05	<0.05	0.055
"	0.10	<0.5	0.08	<0.05	0.060
$CaO + Na_2CO_3$	0.08	<0.5	0.03	<0.05	0.045
"	0.10	0.5	0.045	<0.05	0.055
"	0.23	1.5	0.15	<0.05	0.160
"	0.14	1.5	0.16	0.05	0.156
"	0.23	<0.5	0.16	<0.05	0.095
"	0.20	1.0	0.07	<0.05	0.133
"	0.38	1.5	0.10	<0.05	0.130
"	0.24	1.0	0.075	<0.05	0.140
"	0.135	1.5	0.04	<0.05	0.19
"	0.140	1.5	0.04	<0.05	0.04
"	0.29	2.5	0.065	0.05	0.13
"	0.45	1.0	0.105	<0.05	0.24
"	0.83	1.5	0.13	0.10	0.13

APPENDIX E

LITERATURE REVIEW OF THE HEALTH IMPACTS OF INTERNAL CORROSION

CORROSION IN PREMISE PLUMBING

In 1977, Hudson and Wayner (1) published a report drawing on four case studies documenting water quality deterioration in distribution systems. One case study was of a middle-sized city in the southeastern U.S. which after chlorination, had slightly corrosive water. Circumstances prevailed throughout the distribution system that permitted zero residual chlorine in parts of the system, allowing sulfare-reducing organisms to grow, protected by slimes and corrosion products attached to the pipe walls, while in other parts of the system the residual chlorine concentrations ranged from 0.1 mg/L to 3.0 mg/L.

An average length of household piping of 100 feet one-half inch pipe per metered service was assumed. This corresponded to 2,600,000 feet of small premise plumbing throughout the city, a length slightly greater than in the distribution system. Potential slime concentrations for several pipe sizes, together with the estimated length of pipe and total slime volume reported in the study are presented in Table E-1.

Assuming the same slime thickness in various sizes of pipe, it was noted from the above figures that the potential for water quality impairment is vastly greater in smaller sized pipes than in large pipes. Hence, the greatest potential for water quality deterioration is in premise plumbing, provided there is a source of causative action such as bacteria, or a corrosive water, and provided there is a period of quiescene of sufficient duration for growth to develop.

TABLE E-1. PREDICTED SLIME QUANTITIES FOR VARIOUS PIPE DIAMETERS

Pipe Diameter (inches)	Estimated Length of Pipe (feet)	Potential Slime Quantities	
		Cubic Feet	Volumes per Million
0.5	2,600,000	2,400	500,000
2.0	787,000	2,800	160,000
6.0	1,018,000	12,000	60,000
12.0	221,000	4,900	28,000

Continuing their report, the next issue addressed by Hudson and Wagner was the potential for dislodgement in regard to reversals of flow. Reversals of flow are most likely to occur in the distribution system, whereas they rarely occur in premise plumbing. In addition, with the exception of dead-ends, there is likely to be a continuous movement of water-carrying slime-repressing chloramines through the distribution system; however, in premise plumbing this is an unlikely occurrence. In fact, there will be extended periods of non-movement in premise plumbing, and even more protracted periods when, for example, a residence may be unoccupied due to travel or vacation.

Pipe velocities in water mains are almost always below 7.0 ft/sec, and more commonly below 4.0 ft/sec. On the other hand, a 10 gal/min household draft would produce a velocity of about 10.0 ft/sec. In addition, the smaller size of premise plumbing also makes the hydraulic shear stress near the wall greater than in distribution lines. Thus, not only is the amount of potential slime and corrosion products much greater within premise plumbing, but the potential for dislodgement is greater as well.

Another trouble-causing factor identified but not evaluated quantitatively by Hudson and Wagner was galvanic action. In most distribution systems, the use of insulating connectors is the exception rather than the rule. Nearly all older service lines are galvanized steel pipe. They are commonly connected to cast iron mains via a brass curb stop or ferrule, sometimes with a lead gooseneck, thereby creating a situation where they are three dissimilar galvanic couplings. For example, in household plumbing, the galvanized pipe connects to a bronze case meter in a vault in the yard, which in turn connects to the house and branches into below-floor galvanized lines leading to utility rooms, baths and kitchens. Almost all the risers from the galvanized household service lines to plumbing appurtenances are copper. They connect to brass cold water spigots, to glass-lined steel hot water tanks and to enameled steel dish- and clothes-washers. On the average, each household system contains approximately 10 galvanic couplings. Scott (2) and Wolfe (3) state that certain iron and other nuisance bacteria are "gradient" organisms, that is, they develop where there is a sharp increase in the electrode potential of the system.

Wagner (4) states that many cities ground their electrical systems to premise plumbing. Consequently, with stray currents adding to the factors already mentioned, a bad situation is only made worse. Guerrera (5) in an article addressing this problem states that although the electric industry steadfastly denies that alternating current causes corrosion of metals, it has been demonstrated empirically that corrosion of piping or deterioration of water quality occurs (in areas receiving the same treated water) where the only apparent variable is the presence of stray electric currents. Guerrera advocates the separation of protective grounding electrodes from water piping to protect the integrity of the water supplier's piping system and the quality of the water.

It is evident that greater corrosion and subsequent water quality changes can take place in premise plumbing than in the distribution system. Conditions present in premise plumbing including smaller pipe size, virtually no reversals of flow, extended periods of non-movement, increased pipe velocities, galvanic action, and the grounding of electrical systems exacerbate the corrosion process, especially in the presence of galvanized steel pipe and a corrosive water.

HEALTH IMPACTS OF CORROSION

Microbial Aspects

When high quality finished water leaves the water treatment plant or source and enters the distribution system, microbial changes can occur. Snead, et. al. and Victoreen (7) demonstrated that the greatest deterioration in bacterial quality occurred when chlorine residual was lost. Due to prevailing environmental conditions (i.e., pH, turbidity, trace metal availability, etc.) changes in the microbial population may allow certain microorganisms to predominate including filamentous bacteria or organisms which shed exopolysaccharide fibers, etc., thus creating protected habitats which can act as places of entrapment and harbor items such as corrosion products as well as provide a biological graveyard for cells of bacteria, algae, and diatoms. In these areas where sediments may accumulate, bacteria are often afforded protection from disinfectant action - especially in premise plumbing systems where there are often long standing times and non-existent chlorine residuals (7, 8). In addition, the sediments which can accumulate in these areas provide the organic nutrients to sustain microbial life and encourage selective regrowth of certain bacteria in the microbiota. Several researchers have demonstrated using scanning electron microscopy that bacterial populations are associated with these tubercular incrustations which develop on pipe interiors (8). Where this type of situation prevails, coliform analysis may be inadequate because as the bacterial quality of the total pipe environment changes and regrowth occurs, coliforms may be suppressed or secondary opportunistic pathogens may increase (9). Though inadequately documented as to the potential health risk inherent to the consumer, the regrowth of coliform antagonists and opportunistic pathogens, especially Pseudomonas spp. and Flavobacterium, is of increasing concern (10, 11, 12). These same organisms have been implicated in waterborne infections in various hospital environments.

Pseudomonas aeruginosa is recognized as a human pathogen responsible for fatal septicemias in infants and in adult patients debilitated by burns, malignancies, or old age; as well as responsible for localized infections, e.g., of wounds, the urinary tract, the respiratory tract, the intestine, the eye and the ear (13, 14). The primary etiological agents of these infections are eradicated by antibiotics, and P. aeruginosa can become the new infecting agent. P. aeruginosa cells in these sites serve as foci for hematogenous dissemination. Patients become susceptible to P. aeruginosa infections after prolonged treatment with immunosuppressive agents, corticosteroids, antimetabolites, antibiotics, and radiation. Particularly, with respect to persons with impaired resistance, the isolation of this organism in relatively large numbers from potable waters and in hospitals has been a major cause of concern, especially due to increasing clinical evidence of its resistance to antibiotic therapy. There is presently some evidence which implicates chlorine resistant strains of P. aeruginosa with the recurrence of summer fevers among kidney dialysis patients in the University Hospital. In addition, the presence of P. aeruginosa in surface waters may be related to the high incidence of otitis externa which occurs during the swimming season. Seyfried and Fraser noted a high incidence between such dermatosis and recent swimming activities (15).

As a potential health hazard in the hospital environment, Flavobacterium was reported as a primary pathogen for some surgical patients (16). This organism was isolated from drinking water fountains, sink faucets, humidifying units and portalbe shower heads that retained water and could not be disassembled for thorough cleaning. Flavobacterium can proliferate in such moist environments and can be transmitted throughout all areas of the hospital, including surgical areas, burn clinics, and nurseries.

On community water supply contained Flavobacterium when the standard plate count was less than 10.0/mL and in the presence of chlorine residuals ranging from 0.1 mg/L to 0.8 mg/L (16).

Although no constant relationship appears to exist between the standard plate count and the number of potentially pathogenic invaders that might be present, it is logical to assume that chance occurrences are proportionally greater as the general bacterial population increases.

A study by Seidler et. al. (17) has shown that both water treatment and distribution contribute to the enrichment of multiply antibiotic resistant (MAR) bacteria such as Acinetobacter, Moraxella, Pseudomonas, and Flavobacterium. Site-to-site variations in generic types and differences in the incidences of MAR organisms indicated that shedding of MAR bacteria living in pipelines may contribute to the MAR populations in drinking water. It is believed that these drug-resistant organisms have become more common recently due to the extensive use of antibiotics in medicine and agriculture throughout the world. The highly drug-resistant character of such bacteria could complicate the antibiotic therapy of compromised hosts and also may lead to the regrowth of MAR bacteria in food products and cosmestics which are processed with this water.

It is evident that certain conditions which can prevail in water distribution systems may allow various microorganisms to predominate, such as coliform antagonists and opportunistic pathogens, many of which have also been shown to be multiply antibiotic resistant, and thus create a potential health hazard to the consumer.

Metals Concentrations

Various amounts of metals potentially significant to the health of the consumer can be released into the distribution system depending on the corrosiveness of the water and the type of piping present. In addition, greater metals concentrations are found in standing water, i.e., those waters which are subjected to extended periods of non-movement (1).

In premise plumbing systems subjected to a corrosive water, increased levels of the following potentially toxic metals have been noted: cadmium and lead (primary contaminants), as well as copper, iron, and zinc (secondary contaminants). Lead, copper, iron, and zinc have been observed in concentrations exceeding the EPA maximum contaminant level (MCL) in Seattle distribution standing water.

With respect to at least three of the metals, cadmium, copper, and lead, both national and international evidence associate the problem of corrosive water and an increase in total body intake of these metals. At certain levels, all three are known to have acute health impacts and both lead and cadmium have chronic health impacts. In addition, the less conclusive, but interesting research undertaken by Schroeder and Kraemer (19) and Comstock (20) with respect to corrosive water and cardiovascular mortality make the potential impact of yet another aspect of corrosive water a serious concern of public health officials. There is little information available to implicate any particular waterborne elements in this association (21), but specific hypotheses have been raised and tested relating cardiovascular disease to the toxic metals cadmium and lead (22) and essential metals copper and zinc (23). Whether or not people ingest enough of these elements from their drinking water to affect metabolism is a major unanswered question. Effects of chronic low level exposure to trace metals are not well studied in humans because of difficulties in assessing such exposures. More evidence must be gleaned from carefully controlled and sophisticated evaluations before strong conclusions can be reached with respect to the health impact of corrosive water; referring, in this instance, specifically to the presence of high metals concentrations. This is particularly true with regard to high risk groups such as young, pregnant women and those persons with specific genetic and physiological abnormalities.

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